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Innovative Methods for Emission Inventory Development and Evaluation: Workshop Summary

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ABSTRACT

Emission inventories are an essential tool for evaluating, managing, and regulating air pollution. Refinements and innovations in instruments that measure air pollutants, models that calculate emissions, and techniques for data management and uncertainty assessment are needed to enhance emission inventories. This workshop provided recommendations for improving emission factors, improving emission models, and reducing inventory uncertainty. Communication that increases cooperation between developers and users of inventories is essential. Emission inventories that incorporate these improvements will meet the challenges of the future.

INTRODUCTION

On October 14–17, 2003, NARSTO and the Commission on Environmental Cooperation (CEC) co-hosted a workshop entitled Innovative Methods for Emission-Inventory Development and Evaluation. Some 220 representatives of government, academia, and industry from Canada, Mexico, the United States, and Europe participated in the workshop. Presentations focused on identifying new and innovative tools, techniques, and methods to prepare or evaluate emission inventories with improved quality, cost, and timeliness. In keeping with the NARSTO/CEC North American representation, the workshop centered on emission inventory issues within Canada, the United States, and Mexico. Presentations and discussions focused largely on criteria air pollutants (SO_2, NO_x, O_3, PM, and Pb) and their precursors from both natural and anthropogenic sources; however, techniques discussed at the workshop have applications to air toxics, greenhouse gases, and other types of air pollutants. Both short and long timeframes were considered for characterizing emissions.

The workshop organizers considered the following premises:

- Emission inventories are essential for assessing and managing atmospheric pollution. Emission inventories provide measured data or calculated estimates of pollutants emanating from transportation, industrial, and biogenic sources.
- Uncertainties in emission inventories must be reduced to manage pollution more effectively. Developing and applying innovative techniques for resolving uncertainties will likely enhance and foster more efficient and cost-effective air pollution control.
- Sources of some of the largest emissions as well as those with the greatest uncertainty are either malfunctioning systems or those for which adequate characterization methods are lacking. Cost-effective ways to identify and quantify these emissions require new and/or more intensive methodologies.
- Recent scientific advances suggest innovative techniques that may facilitate future development and evaluation of emission inventories. Application of these methods, in conjunction with

IMPLICATIONS

Emission inventories are the foundation of cost-effective emission control and air quality management strategies. However, emission inventories have often been cited as the weak link in the air quality management process, and have been criticized for not providing products with the desired quality, timeliness, or affordability. Despite successful use in regulatory and other applications, it is essential that emission inventories be enhanced to provide a sound basis for their ever-increasing role in the air quality management process. This workshop provided a forum to address new and innovative tools, techniques, and methodologies that can improve the way emission inventories are developed and evaluated. Recommendations from the workshop should provide momentum for the additional research and development needed to apply these improvements to the emission inventory field. Thus, emission inventories are positioned to be able to meet the challenges of the future.

the more established approaches, could improve the emission inventory development and evaluation process.

To explain these premises and promote linking scientific progress to policy applications, the workshop was framed around three policy questions and three science questions.

The policy questions were:

(1) What policy decisions will be made over the next several years based on current emission inventories?
(2) What is the vulnerability of current approaches affecting these decisions?
(3) What new and innovative techniques can make a difference in these decision processes?

The science questions were:

(1) Do new and more innovative techniques exist on the "technical horizon" that can contribute significantly to emission quantification and are potentially deployable in the foreseeable future?
(2) What are they, what is their nature, and what are their limitations?
(3) How can these methods best be combined with conventional methods?

The science questions were extensively evaluated during the technical presentations at the workshop. Linkage of these scientific issues to the policy questions is illustrated in the summary of policy settings described below. An assessment of the workshop's findings on the policy questions is included in the Conclusion. For additional information, the abstracts, presentation slides, and other materials from the workshop are posted on the NARSTO website (www.cgenv.com).

POLICY DRIVERS

In North America, clean air policy goals are driving the development of new and innovative tools, techniques, and methods for quantifying emissions from air pollution sources. Canada, the United States, and Mexico have special challenges, policy approaches, and timelines, but they share similar goals for improving air quality to protect and improve public health and the environment. Tools, techniques, and experiences gained in one region can be applied or modified for use elsewhere. The following brief descriptions of some of the public health and environmental policy goals in North America demonstrate the growing need to develop new ways of improving air emission inventories, which are the fundamental building blocks upon which planning decisions will be made.

In June 2000, the Canadian Council of Ministers of the Environment (with the exception of Quebec) endorsed the first Canada-wide standards for O_3 and $PM_{2.5}$. The Canadian government set a target of 2010 for achieving these standards. Each endorsing jurisdiction agreed to develop and implement a plan to achieve these standards, and to produce comprehensive reports on progress every 5 years, beginning in 2006. Each jurisdiction also is responsible for identifying situations where natural or background sources, such as forest fires, result in continuing exceeding of the standards despite that jurisdiction's best efforts to reduce pollution from local sources. An improved understanding of air emission inventories in Canada would be important for developing jurisdictional implementation plans, reporting on progress, and demonstrating natural or background influences.

Mexico is currently developing its first comprehensive national air emission inventory for pollutants, including SO_2, NO_x, $PM_{2.5}$, PM_{10}, and NH_3. The initial inventory year is 1999. Periodic updates are anticipated. Mexico faces challenges in locating, quantifying, and speciating emissions from source types such as backyard pottery kilns that are not typically seen in Canada and the United States. It also must develop appropriate methods to quantify emissions from a mobile source fleet unique in fuel quality, driving cycles, and fleet composition. Mexico is in the process of developing and adopting a mandatory emission reporting rule for stationary sources that will require increased knowledge and capacity for estimating and measuring pollution. This effort provides the opportunity to take advantage of new and innovative methods for quantifying emissions.

In the United States, National Ambient Air Quality Standards for O_3 and $PM_{2.5}$, revised in 1997, will force an increasing number of areas to develop implementation plans for achieving these health-based standards. Current schedules anticipate that new areas in non-attainment of the national standards will be designated in 2004. Those areas will have 3 years to prepare State and Tribal Implementation Plans, which include emission control strategies to bring the areas into attainment. Thus, these comprehensive plans are anticipated in 2007. In addition, a national rule to improve visibility in federally protected areas, such as national parks and wildlife refuges, also requires a coordinated effort among all 50 U.S. states and the District of Columbia to achieve national visibility goals by 2064. States will have to develop and submit regional haze plans by 2008 and provide "reasonable progress" reports by 2013 and every 5 years thereafter. With these planning requirements come new challenges to locate and quantify emissions contributing to O_3, $PM_{2.5}$, and regional haze. Efforts are underway to improve the understanding of NH_3 emissions from agriculture and other sources because of NH_3's importance as a precursor to the formation of $(NH_4)_2SO_4$ and NH_4NO_3. Also important are an improved understanding of emissions generated by forest fires and knowledge of sources that contribute to the long-range transport of pollutants from outside planning regions.

STATUS OF CURRENT EMISSION INVENTORIES

Workshop presentations covered source and flux measurements, mobile source characterization, ground, aircraft, and satellite observations, air quality and receptor modeling, emission modeling, and data management and uncertainty assessment. Common to these six areas, emission inventory development and evaluation techniques range between the following two basic approaches: (1) "bottom-up," where emissions are measured or calculated directly by concentration, mass flow, and or stream velocity observations at the source or emissions are calculated on a source-by-source or localized basis; and (2) "top-down," where emissions are inferred by concentration and ancillary measurements in the ambient air downwind from the source or calculations from generalized emission factors and national or regional activity indicators.

Emphasizing the need for innovative development in each of these categories, the organizers stressed the workshop's goal of having its discussions set the stage for continued scientific dialogue and future innovative developments in the field. Institutional schedules and limitations often require emission inventory developers to rely on conventional methodology, and leave little motivation to consider or apply non-routine applications. The workshop was intended to help provide a break in this routine. Accordingly, the workshop's organizers encouraged all attendees to apply their creative skills and "think outside of the box" during participation in the sessions.

Three overarching features that emerged during discussions throughout the six technical sessions are especially of note. First, many of the current comparisons of emissions using top-down (e.g., aircraft measurements of concentration fields) and bottom-up techniques (e.g., from traditional emission inventories) show large discrepancies. This indicates that annual average emission inventories developed for regional assessments may not be adequate for comparison with observations over a short period of time for a specific geographic area. Second, emission inventories applied for chemical transport modeling applications usually require much higher temporal and spatial resolution than is available in conventional inventory data sets. Both of these features imply that a strong need exists for more innovative measurement, estimation methodologies, and data management techniques to resolve these issues. The third noteworthy feature is that a significant amount of research and development is under way in the emission measurement, characterization, and inventory fields. Although much of this work is not revolutionary, it can provide meaningful enhancement to the quality, timeliness, and cost-effectiveness of emission inventories.

The technical content of the six technical sessions is summarized below. Because this information is by necessity highly condensed, the reader is encouraged to consult the detailed presentation material for the workshop, which can be accessed on the NARSTO website (www.cgenv.com/narsto). Recommendations from individual discussion sessions are summarized at the end of this paper.

Source and Flux Measurement

Within the context of the workshop, "flux measurements" pertain to direct or indirect measurements of pollution fluxes (amount of pollution issuing from a unit area of the Earth's surface per unit time), whereas "source measurements" generally refer to direct measurements of pollutant quantities (amount of pollution per unit time) issuing from specific source locations. Typical flux estimation techniques include chamber measurements, vertical profile measurements, and correlated measurements of pollutant fluctuations. Discussions covered flux and source measurement studies directed at volatile organic compounds (VOCs) from industrial, urban, and biogenic sources, as well as $PM_{2.5}$, NH_3, fugitive dust, and visibility reducing species.

VOC Emissions from Refineries, Chemical Plants, and Urban Areas. Several new source measurement techniques employed during the Texas Air Quality Study in the summer of 2000 (TexAQS 2000)[1] showed that O_3 and VOC concentrations in the Houston harbor refinery areas were greater than could be accounted for by the reported state and local emission inventory. Photochemical air quality modeling inputs using underestimated VOC emission rates underpredict O_3 production in the Houston area. To improve modeling results, ratios of VOCs to NO_x emissions were adjusted. The collection of speciated, high time resolution ambient VOC and NO_x data is costly, and may be limited to O_3 non-attainment areas deploying specialized ambient monitoring such as photochemical assessment monitoring stations. Detailed ambient data are, nevertheless, an effective means to guide adjustment of air quality modeling results, which serve to augment current emission inventory methods.

In Houston, concerted efforts at chemical plants and refineries to track down high VOC emissions focused on industrial flares, cooling tower emissions, and fugitive VOC leaks. Quantifying elevated flare emissions was found to be a complex problem because of the hundreds of process vents being routed to the flares. Ground-based passive Fourier transform infrared spectroscopy is being tested to obtain mass and speciated emission rates and accurate flare combustion and VOC destruction efficiencies over a range of operating conditions. This technology is in the early phases of development, and it is expected to be several years before new emission factors from this method become available. Meanwhile, improved emission estimates could be achieved if flare destruction efficiency is provided with greater specificity.

An improved method has been proposed for estimating cooling tower VOC emissions based upon the concentration of "strippable VOC" in the cooling water, the water circulation rate, and the identification of process fluid leaks into the cooling water. This approach is believed to give much more accurate emission estimates than those currently determined by using U.S. Environmental Protection Agency (EPA) default emission factors.[3] This approach is a near-term improvement that is inexpensive to implement and requires no policy changes.

Optical imaging instruments have been developed that detect and display VOC leaks. The portable units provide a more cost-effective method of detecting fugitive emissions in chemical plants and refineries than conventional leak detection and repair using currently approved EPA methods. The result is significantly improved detection of leaking equipment and piping. Several published field studies have been conducted, and more are underway to evaluate active laser and passive infrared systems. A long-range active laser camera suitable for remote sensing of fugitive emissions is commercially available. An EPA rule allowing the use of gas imaging is expected in 2004.

VOC emissions from an entire refinery, or even a small urban area, can be measured by using differential absorption light detection and ranging (DIAL LIDAR or DIAL). DIAL measures differences in absorption of UV and IR light by using pulses of laser light, a transmitter (laser), a receiver (telescope), and a detector. DIAL generates three-dimensional plots displaying VOC concentrations and fluxes throughout refineries and industrial plants. DIAL measurements are required at many refineries in Europe as a complementary technique to leak detection and repair. DIAL can provide more accurate emissions data than other standard estimating techniques for petrochemical and other facilities. On a larger scale, aircraft DIAL measurements can be used for metropolitan and regional areas, which suggests a number of intriguing applications for evaluating emission inventories.

Biogenic VOC Flux Measurements. Biogenic VOC (BVOC) emissions are important in determining the chemical composition of the atmosphere and for estimating O_3 and secondary particulate production. Considerable advancements in methods on all scales have been made in the past decade. BVOC flux measurements are made over leaf-scale (enclosure measurements), canopy-scale (above-canopy tower measurements), landscape-scale (tethered balloon), and regional-scale (aircraft and satellite measurements) observations. Seasonal and diurnal variations characterized with eddy flux measurements from above-canopy towers are used to evaluate canopy-scale emission models.

Eddy covariance (EC) flux methods and fast-response chemical sensors have matured to the point where it is now possible to make direct emission measurements of a range of VOCs. Disjunct EC provides further flexibility in coupling a variety of chemical sampling methods or sensors to turbulence measurements to yield direct emission flux measurements. EC and disjunct EC methods directly measure VOC fluxes from forested environments, and have the potential to yield similar data for urban environments. The source footprints from EC and disjunct EC methods generally extend from a few hundred meters to several kilometers and, as such, the data can provide a basis for direct evaluation of emission models and/or gridded emission inventories.

Aircraft-based methodologies are being developed for measuring the rate of exchange of BVOCs with environmental surfaces. EC measurements made from an aircraft platform are capable of discerning eddy patterns of turbulent BVOC flow above a forest canopy. The measurement approach uses a variety of eddy sampling techniques in concert with fast- and slow-response chemical and turbulence sensors to determine air–surface exchange rates. Continued development of sensors will require a fast-response global positioning system and an upgraded data system.

$PM_{2.5}$, NH_3, Fugitive Dust, and Visibility. New $PM_{2.5}$ emission factors and speciation profiles are needed for combustion sources in the oil, gas, and power generation industries. Characterizing $PM_{2.5}$ emissions from combustion systems is difficult because the nucleation, condensation, and coagulation processes that occur upon dilution in the atmosphere change the amount, size, and composition of PM emissions. Dilution sampling is a technique that simulates the effects of rapid cooling and dilution on PM emissions from combustion systems. A new dilution sampler has been designed to better simulate the effects of rapid cooling and dilution on fine PM emissions from combustion systems. The sampler is configured to collect the PM component species as well as the emission mass, providing results directly comparable to that of ambient $PM_{2.5}$ speciation samplers. Field tests on several full-scale gas and residual oil-fired sources have been conducted to characterize mass emissions and composition. The initial indication is that the emission factors

derived from dilution sampling are notably less than those estimated from the EPA's default emission factor method.[3] An American Society of Testing materials task group is being formed to address the development of a protocol for the dilution sampling technique. With adequate funding, a minimum of 3–4 years will probably be needed to refine the technique and complete the method approval process.

A significant fraction of ambient $PM_{2.5}$ is secondary NH_4NO_3 and $(NH_4)_2SO_4$. To model the formation of these species, an accurate NH_3 inventory is needed. Agricultural livestock is the largest single source of NH_3, but measuring its emissions represents an enormous task. Passive samplers have been shown to achieve measurement accuracy similar to active flux samplers and meteorological data. This approach has the potential advantage of economically compiling a broad inventory base for ambient NH_3 emissions. An inexpensive fabric denuder has been designed for the passive NH_3 flux measurement. NH_3 flux data can be integrated over the extent of the source to determine "area source" emission rates.

Primary $PM_{2.5}$ emission sources include wind-blown fugitive dust. The Portable In-Site Wind Erosion Laboratory (PI-SWIRL) is an instrument designed to determine fugitive dust emission factors from soil surfaces; it consists of an open-bottom cylinder containing a horizontal blade that rotates several centimeters above the soil surface. The blade rotation induces a shear stress above the test surface, simulating the stresses caused by high winds. Though not as accurate as some of the larger field wind tunnels, the PI-SWIRL is an easily deployed field unit capable of making multiple measurements. Before results can be accepted, the formal process of developing a sampling protocol to provide acceptable emission factors will be needed. Development of the current prototype and an optimized version could be completed within 2–3 years. The potential to make hundreds of measurements in a relatively short time can assist in developing more accurate models for emission factors.

Mobile Source Characterization

The development of an accurate mobile source emission inventory requires cost-effective ways of determining emission factors and activity indicators for a representative sample of in-use vehicles operated over a broad range of operating conditions, fuels, and ambient conditions with high temporal and spatial resolution. The data must be collected in a manner that enables its use in an emission inventory model that is capable of providing output to a broad range of users. An in-depth review of current research activities in this area is available in the summary of the Thirteenth CRC On-Road Vehicle Emissions Workshop.[2]

A large fraction of the HC, CO, and NO_x emissions from the light-duty vehicle (LDV) fleet comes from a small percentage of vehicles. Many of these high emitters are older vehicles that have been poorly maintained or modified. Recent studies of PM emissions from gasoline LDVs also show that a few high emitters dominate the total PM emissions. Acquisition of emission distribution data is difficult largely because recruiting vehicles for emission testing in an unbiased manner is problematic, as is testing a large enough sample of the in-use fleet to ensure representative emissions.

The desire for increased fuel economy in the LDV fleet may cause a large increase in the number of gasoline hybrid vehicles and diesels. This will also add to the complexity of the emission inventory. Given these issues, the development of improved mobile source emission measurement techniques is expected to remain very important.

Strict exhaust emission regulations for on-road, heavy-duty vehicles (HDVs) and nonroad vehicles have lagged those for LDVs. However, new regulations for these categories are being phased in; hence, in the future, HDV sources will also have a diverse in-use fleet that is heavily impacted by a small number of high emitters. The importance of heavy-duty diesels has grown, as evident in the fact that they now account for about half of on-road NO_x emissions.

Mobile Source Emission Measurement Methods. Exhaust emission measurement methods include laboratory studies using chassis or engine dynamometers and emission benches. This methodology is the gold standard for emission testing because it has high accuracy and precision. Unfortunately, laboratory testing is time consuming and expensive; thus, it is usually not used for large numbers of in-use vehicles. Continuous improvements in laboratory testing are mostly focused on increased sensitivity for measuring emissions from low-emission vehicles and improved analytical instruments for measuring unregulated emissions. Measuring PM emission rates from low-emission vehicles is receiving considerable attention, as is the physical characterization of the PM. Several particle mass, size, and surface area instruments have been developed and are being tested. Hence, standardized methods and instrumentation can be expected in a few years.

Transportable dynamometers have been used for both on-road LDVs and HDVs, thereby eliminating the need to transport vehicles to a laboratory. However, costs remain high and alternative solutions for obtaining in-use data are being sought. Enhanced inspection/maintenance (IM) programs test vehicles on a chassis dynamometer by using the standard IM240 driving cycle. This is a useful source of data for inventory work. It is hoped that the on-board diagnostic systems required in newer vehicles will eventually supplant the need for IM programs. Hence, the data from dynamometers is unlikely to expand.

Evaporative emissions are a major contributor to mobile source VOC emissions. With the exception of refueling emissions, these are measured in the laboratory by using enclosure techniques. These procedures are time-consuming and expensive; hence, there are relatively few in-use evaporative emission data. There do not appear to be any new and improved methods for measuring evaporative emissions. This area needs attention in future studies.

Roadway tunnel studies have been identified as an effective means of determining fleet emission rates of in-use vehicles. Improved analytical instrumentation has facilitated the expansion of the number of species characterized in these studies to include air toxics, greenhouse gases, particulates, and ozone precursors (NOx and speciated hydrocarbons), as well as the regulated emissions. Limitations of roadway tunnel studies include the lack of individual vehicle data, lack of suitable tunnels, limited driving conditions, and the need to separate diesel from gasoline vehicle emissions. Emission rates from tunnel studies have been used effectively in evaluating portions of the emission inventory models, tracking year-to-year trends in fleet emissions, and in providing data for fuel-based inventory models. This is a mature methodology that should continue to be used, but it will not provide the data needed for detailed emission inventory models based on specific vehicle data over a broad driving activity range.

Ambient measurements in mobile source-dominated atmospheres are also used. When performed with long-path techniques at roadside, they are essentially tunnel studies without enclosure walls. Total flux measurements are rarely made in these studies. Hence, it is not possible to generate emission rates in the traditional grams-per-mile units used in regulatory emission factor models. Instead, emissions are usually ratioed to CO_2 or another major exhaust species. If CO_2, CO, and VOC are measured, then measurements can be put on a fuel-consumed basis. This is especially useful for fuel-based inventory models.

A special case where ambient measurements currently are important is the study of exhaust nanoparticles. Formation of these particles is highly dependent on dilution conditions, which are extremely difficult to simulate in the laboratory. Hence, ambient studies are needed to compare to laboratory measurements. Another special ambient case discussed was the measurement of plumes from marine vessels currently underway. Initial results affirmed standard methods,[4] but more work in this area is needed.

High-speed analytical instrumentation can be used in vehicle chase studies. These studies can obtain emission data from selected vehicles, although they don't obtain as much detail about vehicle operating condition as do the portable emission measurement system (PEMS) studies discussed below. Emission data are fuel-based, because total flux measurements are not possible. Cost and availability have limited the use of this instrumentation. Continued development of instruments will make this more practical.

Entire conventional emission measurement systems have been placed in trailers to be driven by heavy duty diesels. Because exhaust flow is measured, gram-per-mile emission rates can be determined. Though very valuable, this is an expensive approach to obtaining in-use test data from a large number of vehicles. An instrumented trailer has also been used to measure the emission rate of entrained road dust, a major PM source.

As commonly employed, remote sensing uses a cross-road optical beam to measure emissions from passing vehicles. Measured species are ratioed to CO_2 concentrations. Hence, it is also a fuel-based measurement. The advantage is that thousands of vehicles can be measured in a single day. The disadvantage is that only 1 s of data is obtained for each vehicle. Remote sensing data have been used to generate fuel-based emission inventories, to evaluate existing inventory models, to track trends in on-road emissions, and to assess IM programs. Vehicle speed and acceleration are generally measured to put the emissions on a vehicle-specific power basis. It has been shown that vehicle-specific power can be used to predict fuel economy, thereby raising the possibility that remote sensing data can be put on a grams-per-mile basis. The ability to detect interference from condensed water plumes has recently been added. Laser systems that have been used on a research basis are able to greatly expand the number of species measured. Several organizations are working on adding PM measurement capability to remote sensing. These advances in remote sensing should greatly enhance its use in the future.

Of all the mobile source emission measurement techniques discussed, PEMS and portable activity measurement systems (PAMS) have made the most progress in the last few years. These systems are used on a vehicle to measure real-time emission rates or activity parameters for several hours. The systems plug into the on-board diagnostic computer and thereby obtain a great deal of vehicle operating information. They also use global positioning to track vehicle location. Reduced size, increased speed of installation, and ruggedization all contribute to the ease of use of these systems. Most effort for the commercial systems has been put into measuring regulated emissions, although a variety of research instruments have shown that other measurements are practical. The biggest element lacking on current systems is accurate PM mass measurement. Work is underway to solve this problem. It is anticipated that use of these instrument packages will greatly increase the amount of in-use data available for both on-and non-road vehicles. The EPA's Motor Vehicle Emission Simulator (MOVES) model is being designed with this in mind. It was also noted that aftermarket instrumentation can be added to a vehicle that monitors the on-board diagnostic system to identify emission problems and report them to a central location. This has been discussed as an addition to IM programs, but is unlikely to have broad enough use for emission inventory purposes.

Mobile Source Emission Models. Two emission factor models were discussed. The EPA's national mobile inventory model combines the MOBILE6 and NONROAD models with a national county database. The graphical user interface makes it easy to create inventories. The model, which is scheduled for release in 2004, is not viewed as a replacement for either MOBILE6 or NONROAD. MOVES is the EPA's new mobile source inventory model. Unlike MOBILE6, MOVES will group emission data based on vehicle-specific power and speed, thereby making it possible to model any driving pattern. Modeling can be done from the microscale to the macroscale. PEMS- and PAMS-based studies are expected to be the major source of the data needed to support the model. Uncertainty estimates will be included in the model. In addition, the model is being designed to integrate with the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation model, Argonne National Laboratories' well-to-wheels life-cycle inventory model. The first version of MOVES will cover on-road greenhouse gas emissions, and is scheduled for release in 2005. Aircraft, commercial marine, and railroad emissions will be added in 2005, and a full on-road vehicle version with all the criteria pollutants is expected to be available at the end of 2005. Other nonroad emissions will be added in 2006. If successful, this will be a major advance in mobile source inventory modeling and emission estimation.

Model validation efforts need to continue, for example, to resolve problems with CO predictions in MOBILE6. MOBILE6 will continue to be used for several years, and many of its methodologies will be included in MOVES. Modified versions of MOBILE6 are used in Canada and Mexico. Hence, understanding current sources of error should help both current and future modeling efforts across the continent.

Fuel-based models are also used; these models use fuel-based emission rates, such as those obtained in remote sensing, tunnel, and chase studies, and fuel use data. The fuel use data are based on fuel sales for a given area or region. Fuel sales are monitored closely. Hence, fuel data are more accurate than the vehicle-miles-traveled estimates needed for activity-based modeling. Fuel-based modeling cannot be used at the microscale, because the fuel consumption at any given location is not known. The models can, however, provide valuable insights regarding emissions over long time periods and large spatial scales. Thus, their primary value is in developing urban to regional scale inventories and comparing those to the more traditional inventory.

Ground, Aircraft, and Satellite Observations

Ambient concentration measurements at the ground and aloft, combined with spectral imaging by satellite or aircraft, are useful means of verifying elements of emission inventories as well as the location or extent of air contamination from sources. Ambient concentration data have been used extensively in calculating chemical mass balances with source profiles and in performing statistical analyses to evaluate emission estimates. Photography and images from remote sensing have been used to identify plumes or regional haze elements for a number of years. The presentations in this session provided a perspective on recent innovative advances in using ambient data of various types for top-down approaches to testing the veracity of emission inventories. The latter are based on conventional bottom-up assemblies of emissions at different geopolitical levels using emission factors, activity patterns, and emission control information by source.

The techniques presented focused on refinements in the use of ground data, aircraft tracking with fast-response instruments, and combining source-based models with receptor techniques. In addition, an in-depth discussion of the state of knowledge was presented about applications of satellite data for source identification.

Ambient Data for Sources. Historical air monitoring data implicitly contain information about trends in emissions over many years. Ambient trend data can be compared with emission inventory trend data to evaluate consistency. An example of such a comparative history was given for CO. The decline in ambient CO concentrations from 1985 through 1999 in the United States was compared with reported CO emissions in urban and rural areas. The annual decrease in ambient concentrations is a factor of 2+ higher than the decrease in emission trends reported in EPA inventories. To help remove meteorological variables, trends in concentration ratios are frequently examined. For example, the CO/NO_x emission and ambient concentration ratios are dominated by vehicular emissions in cities. This ratio declined by a factor of 3 between 1987 and 1999, a reduction about twice that reported in mobile source emission inventories. These results suggest that the rate of decrease in U.S. CO emissions is underestimated in current inventories.

Routine particulate carbon measurements have recently been used to infer emissions from biomass burning in the southeastern United States. Using the Southeastern Aerosol Research and Characterization Study measurement network data, it was possible to estimate emission factors for vegetative burning where fires could be identified systematically from continuous ambient concentration data for black and organic carbon combined with CO and NO_x. Tracing urban and point source plumes using ratios of NO_x, CO, and SO_2 concentrations, combined with particulate carbon data, provided the means of separating biomass-burning sources from other sources.

Combining Air Chemistry and Meteorological Data. A "classical" method for tracing sources employs a combination of ambient concentration observations with wind direction and speed and mixing height. Using direct correlation between wind direction and changes in concentration along with air quality models, one can infer the emission rate and location of major sources. Examples of this approach, using semicontinuous observations of PM mass concentrations and composition, were reported for the New Mexico–Mexico border area. Interpretation of the diurnal changes in particle concentrations combined with Pb and Sb concentrations and carbon data enabled identification of local sources on both sides of the international border.

The application of fast-response instruments for measuring gas and particle concentrations with specified uncertainty has greatly enhanced the ability to apply air chemistry meteorological analysis for quantitative source characterization. The high temporal and spatial measurement resolution offers major advantages in the tracking of emitted pollutants from sources.

Aircraft-based tracking of gases in plumes from power plants, refineries, and other industrial sources has been used to determine emission rates. Emission rates can be determined to $\pm 20\%$ by using emission ratios of SO_2, NOx, CO, CO_2, and VOC. Results are consistent with source continuous emission monitoring (CEM) data, but differ significantly from estimates based on generic calculations, for example, from annual inventory estimates.

Highly time-resolved data from the Maryland–EPA ambient air research site (Baltimore Supersite) have been used to quantify emissions from nearby sources. Dispersion modeling using particle and metal concentration data, together with meteorological data, was used to identify coke oven emissions and emissions from a local power plant. A similar approach to source emission estimation in the Tampa, Florida, area used fast-response, ground-based monitoring of gas and particle concentrations to characterize emissions from fertilizer manufacturing and coal-fired power plants.

Time-resolved measurements with fast-response instrumentation also can be used effectively for specialized diffuse source studies by making surface flux measurements. A multi-tower system has been used for measuring particulate fluxes of fugitive dust under different meteorological conditions. Measurements of PM_{10}, $PM_{2.5}$, and PM_1 have been made with this system, thereby providing unique data in a sparsely vegetated environment for dust flux in different particle sizes with and without the presence of vehicles on a nearby unpaved road. Such data provide badly needed new knowledge quantifying short-term fluxes of dust from the ground.

Remote Sensing and Satellite Observations. For more than two decades, scientists have hoped that satellite remote sensing would reach a stage where gas and particle concentrations or column burden could reveal information about large-scale source contributions. The current satellite fleet has obtained a variety of data that characterize key chemical components of the atmosphere. These are summarized in Table 1. The array of satellite data collection is constrained in (vertical and horizontal) spatial resolution and is limited to latitudes below ~65°, with minimum spatial resolution of ~20 × 20 km. Most of the data are "column" average burdens that require interpretation in terms of ground level or other sources. However, the collection of sustained data over a period of many years offers a unique opportunity for source evaluation on a sub-continental and larger scale. This is important for planetary scale issues such as climate response to gas or aerosol particle forcing. Satellite data and imaging are now being used to obtain knowledge of a variety of sources such as industrial activity, urban emissions, biogenic emissions, and fugitive dusts and fires.

Most of the work identifying sources with satellite remote sensing had been of a qualitative nature until recently. Qualitative studies have included the use of remote sensing to do the following: determine vegetation patterns and land use, along with photosynthetically active radiation for use in estimating biogenic sources; improve on knowledge of fire source potential by the U.S. Forest Service; and to characterize the long-range transport of particles from optical depth changes and haze parameters on a sub-continental scale associated with large fires across North and South America and Africa. Fires are readily identified in the IR from heat generated from combustion. Aircraft imaging has also been used for qualitative source identification, such as identifying dust

Table 1. Atmospheric chemistry measurements from space (NASA, 2003).

Sensor[a] Species	TOMS	GOME	MOPITT	MODIS	SCIA-MACHY	TES	OMI	HRLDS	MIPAS	SAGE III	MLS
Launch	1979	1995	1999	1999	2002	2004	2004	2004	2001	2003	2004
O_3	Col[b]	Col			Col/L	Nadir[c]/L	Col	Limb[d]	Limb	Limb	Limb
CO			Nadir		Col/L	Nadir/L			Limb		Limb
NO_2		Col			Col/L		Col				
CH_2O		Col			Col/L		Col				
SO_2		Col			Col	Col	Col				
H_2O					Col/L	Nadir/L		Limb	Limb	Limb	Limb
NO						Limb					
HNO_3						Limb		Limb	Limb		
CH_4			Nadir		Col/L	Col		Limb			
CO_2					Col/L						
BrO		Col			Col		Col				
HCN											Limb
Aerosol Particles (Optical Depth)	Col			Col	Col/L		Col	Limb		Limb	

[a]Acronyms for instrument packages on different satellite platforms, launched at different times since 1979; [b]Col, total atmospheric column from surface to space; [c]Nadir, column measurement with distinction of some vertical layers; [d]Limb (L), scan through atmosphere with vertical resolution to the tropopause.

and particle sources in the West to improve emission inventories of fugitive dusts.

Perhaps the most intriguing work with satellite data is the attempt to quantify NO_x and BVOC emissions. NO_x emissions have been estimated a posteriori from the 1996–1997 Global Ozone Monitoring Experiment satellite NO_2 column data, and the GEOS-CHEM model. The derived monthly NOx emissions were estimated to be within a 50% error. Geographical differentiation showed good correlation between the two methods, but there were significant differences between conventionally derived estimates and the satellite-derived results in some locations. Another study analyzed the1995–2001 formaldehyde (CH_2O) data from the Global Ozone Monitoring Experiment to infer column BVOC concentrations. During the vegetation-growing season, global large-scale CH_2O is dominated by isoprene photochemical oxidation. This top-down estimate of isoprene emissions is generally similar to that estimated by the Global Emission Inventory Activity or Biogenic Emission Inventory System, including interannual variability driven by leaf area index. Improvement of the satellite-based method will depend on obtaining new data for CH_2O formation from the oxidation of VOCs under different atmospheric conditions.

Air Quality and Receptor Modeling

Several modeling approaches are being pursued for emission inventory improvement, both in terms of direct measurement and reporting as well as in validating inventory figures. The modeling tools include the use of receptor modeling-related techniques to evaluate and improve inputs to inventories. A range of these developments was described at the workshop, and the steps needed to put them into more widespread practice were examined. The developments may be placed in the following five groups: (1) empirical methods to assess the relative contribution of emission sources to air concentrations at a site; (2) top-down estimation by inverse modeling; (3) model-based sensitivity analyses to determine the importance of inventory changes suggested by measurement discrepancies or missing sources; (4) improved modeling assistance tools such as activity patterns, temporal variability information, and regional input information; and (5) conceptual models used to guide air quality analyses.

Emission inventory values are subject to many uncertainties resulting from the wide range of information sources, approximations, and empirical models used in their compilation. Consequently, it is useful to compare emission inventory values with emission estimates derived from independent approaches. Receptor modeling combined with detailed ambient measurement data and positive matrix factorization analyses can be used to identify the sources and temporal variability of emissions when chemical speciation profiles are known. This powerful approach informs modelers and emission inventory compilers as to which emission sources are contributing to air concentrations and which need more attention, both with respect to specific sources and source categories. Examples of this technique are most successful when there is a rich, speciated ambient measurement database, such as those obtained in the TexAQS 2000 study.[1] The cost of establishing and maintaining an intensive monitoring network is the principal drawback.

Recently, inverse modeling has been used in regional-scale air quality modeling to investigate suspected high

NH_3 values in emission inventories. Inverse modeling is applied where linearity is assumed to exist between emissions and modeled responses—in the case of NH_3, between emissions and the modeled wet deposition of NH_4^+. Results of model sensitivity studies varying the amount and temporal variation of NH_3 emissions are compared with spatial and temporal patterns in monitoring data. This technique has proven important in guiding research efforts to improve NH_3 emission inventory components. The power of inverse modeling is improved by the presence of a dense ambient measurement network. Although experience in regional application and modeling accuracy is limited at this time, as inverse modeling techniques improve they may allow top-down estimation and adjustment of emission inventories.

As noted previously, ambient air quality measurements, especially from high spatial and temporal resolution networks, have proven effective as a validation tool for emission inventories used as input to regional air quality models. If model results significantly over- or under-predict ambient concentrations relative to monitoring data, improvements and/or adjustments in inventory components may be suggested. Discrepancies, particularly those during midday hours, may infer inaccurate emission inventory values for specific sources, source categories, or missing sources. The drawback of this approach is the cost of intensive ambient measurement networks, and the limited degree to which region-specific information can be applied elsewhere.

Emission inventories can benefit from the improvement and application of a variety of supporting tools and techniques. These include evaluation of monitoring and modeling data statistics, better representation of emission model input information, and region-specific studies and information. For example, ratios of monitored components of emissions may be used to identify the source categories and relative amounts of emissions (e.g., $^{12}C/^{13}C$ or $^{12}C/^{14}C$ ratios) compared to the emissions accounted for by an emission inventory. Improved knowledge of the diurnal, weekly, and seasonal temporal variability of the emissions of different source categories from field studies can make an appreciable difference in emission inventories and in temporal allocation of emission values for modeling. Temporal variability of emissions may also vary regionally (spatially); mobile source patterns are a primary example. In some cases, local or regional studies of direct or indirect indicators of emissions or the effects of emissions, such as metals deposited to the soil, can also provide valuable guidance to improving or interpreting emission inventories for limited areas. Emission inventories typically characterize values as annual numbers. In addition to spatial and temporal variability information needed for modeling, the uncertainty and temporal variability inherent in emission values is better described in terms of stochastic representations than single numbers. Stochastic representation is at variance with the regulatory need for fixed numbers, but does provide some needed indication of uncertainty or reliability.

Finally, conceptual models based on laboratory or basic "box-models" may be used as analytical guides to the improvement of emission inventories. For example, fractional aerosol coefficients have been applied to precursors of secondary organic aerosols to estimate the contribution of source categories to the formation of secondary organic aerosols.

Emission Modeling

Emission models provide the interface between emission inventories and air quality simulation models, matching inventory data to temporal profiles, speciation profiles, and geographic locations. In addition, emission models are used to calculate emissions and develop air quality model-ready inventories for mobile sources (on- and off-road), biogenic emissions, NH_3 emissions, and other sources. Improvements in emission models can improve air quality modeling results and thereby provide stronger support for policy decisions. At the workshop, the following advances in emission modeling were discussed: (1) using air quality measurements to improve inventories; (2) introducing variability into inventories with probability models; (3) using CEM data to improve emission characterization; (4) enhancing models for biogenic emissions, thereby improving inventories for Mexico; (5) improving models for specific contaminants (including NH_3, carbonaceous aerosols, and SO_2 emissions); and (6) developing a new emission model paradigm.

Performance Evaluation. The reconciliation of emission measurements made during TexAQS 2000 with the emission inventory available for the region showed that, despite the quality of the inventory for the area, monitoring data provided new information that led to important improvements in the inventory. These inventory improvements had significant consequences for O_3 control measures that are estimated to have saved some companies millions of dollars in potential control costs while at the same time focusing on the controls that were most needed.

Incorporating Probability. Further intensive analysis of the inventory, source-specific operating information, air quality measurements, and modeling results emerging from TexAQS 2000 led to the conclusion that industrial emissions of highly reactive VOC species were inadequately represented in the annual inventory when it was used for episodic application. In addition, significant spatial and temporal variability was identified in VOC emissions from major industrial sources such as petrochemical facilities that could not be expected to be captured in the annual inventory.

To incorporate variability into emission estimates, researchers analyzed operating records and developed probability distributions to represent various scales of variability. The challenge now is to incorporate these estimates into inventories and modeling analyses. As an example of how probabilistic or stochastic emission estimates may be used in modeling, repeated air quality modeling runs were used to quantify the effect of this variability in emissions on ambient concentrations. Simplified sub-domain models were used to minimize modeling costs. The sub-domain models were calibrated to represent the region of interest by using an initial run of a regional grid model to develop boundary conditions and establish model parameters.

Using CEM Data. The EPA has been using hourly emission data, gathered as part of the U.S. Acid Rain Program, to create an annual emission estimate for SO_2 and NO_x in the National Emission Inventory. For modeling inventories, hourly data should be included in more detail, but there are several challenges to accomplishing this. Experience in Texas indicates the importance of allowing affected sources to review the data. The biggest difficulty is in matching records from the Acid Rain Program inventory and the National Emission Inventory, because common identifiers are not used and duplicate and missing records must be addressed. Standardizing source identifiers and sharing software and methods for incorporating the Acid Rain Program data would be useful for upcoming state implementation plan modeling for O_3 and PM.

Specific Contaminants. Emissions of biogenic species are calculated by using emission models such as the EPA's Biogenic Emission Inventory System model and the Glo Biogenic Emission Inventory System model used by the State of Texas. As agencies prepare to analyze 8-hr O_3 problems and issues related to PM and atmospheric aerosols, there is a need to improve estimates of biogenic emissions, including episodic emissions from fires. Estimates show wildfire emissions are significant contributors to ozone and PM, and estimating fire emissions will be important to baseline models and projection models. Improved emission factors are needed for biogenic species important in all locations. Areas of Mexico are certainly candidates for additional research on this topic. Also, PM and semi-volatile BVOC species are becoming more important and need more thorough characterization.

The five U.S. regional planning organizations are coordinating the development of a model that provides a process basis for temporal allocation of ammonia emissions. Improving estimates of agricultural NH_3 is important because of their large emissions as well as the large uncertainties in their estimates.

Significant concerns about previous estimates of black carbon are important for global climate modeling in addition to ambient PM modeling. New estimates emphasize the importance of residential coal and biomass combustion internationally. Emission sources differ significantly between developing and developed regions. Standardized measurement and estimation methods are needed for black carbon that go beyond the current thermo-optical filter-based techniques, especially because this pollutant is not now included in conventional inventories.

Evaluation and Uncertainty Assessment and Data Management

Performance evaluation of emission inventories or models has been addressed with various methods, but an acceptable formalized approach has not been adopted, even though ambient concentration data or air quality modeling has been used informally. As noted above, the intercomparisons between ambient data and air quality modeling results have led to substantial improvements in some emission estimates. The accuracy of emissions from some source categories, such as motor vehicles, chemical plants, or refineries, has come under scrutiny when control measures for sources in these categories were proposed as a part of implementation plan development.

Evaluation and Uncertainty Assessment. Statistical methods have been used to evaluate emissions in reference to other "ground truth" (e.g., comparison of historical VOC/NO_x ratios from emission inventories with air quality monitoring data). In the past, the ratios from some species in the emission inventories were lower than those determined by ambient monitoring. This has led to "proportional" adjustments in emission inventory estimation methods. More recent data show these ratios to be in closer agreement with monitoring data and, hence, model performance has improved. The process was the following: (1) use top-down analyses such as ratio comparisons, (2) evaluate air quality modeling, and (3) refine the conceptual model based on these results. A recent example is the determination that, in some cities, emissions on weekends differ substantially from those on weekdays.

The model/emission VOC/NO_x ratios in Houston were examined in the TexAQS 2000 field study.[1]

Also included were analyses of ratios of specific VOCs to NO_x and a wind direction component. These analyses indicated that the routine emission estimates for nonpoint sources are consistent with ambient measurements. However, the routine emission estimates for point sources appeared to be significantly lower than the measurements indicated, suggesting that the point source emissions for highly reactive VOCs should be increased. Statistical approaches for estimating the uncertainty in power plant NO_x and SO_2 emissions show promise, but require further development.

The uncertainty in methods used to determine biogenic emissions was addressed by comparing biogenic emission data with data from airborne sampling conducted during TexAQS 2000. Results showed the sensitivity of emissions to ground cover, air temperature, and cloud cover. Another study addressed changes in biogenic emissions when different sources were used for air temperature and solar radiation data. In both cases, it was shown that significant differences in emission estimates would result if different data sets were used.

An evaluation and inventory adjustment technique was used for a large area in Asia based on integrating emission inventory, air quality modeling, and ambient monitoring data. The emission inventory was developed by using the best available emission data. Air quality models were used to predict concentration profiles, which were used to establish flight paths for airborne sampling. Monitoring results were compared with the emission-generated estimates by using ratio methods and back trajectories. Then, adjustments were made to refine the emission inventory to match ambient observations.

Data Management. A new user-friendly system has been implemented in the South Coast Air Quality Management District to develop its emission inventory. This computerized system is used by the facilities to report their emissions to the District. The design and development of an integrated permit and inventory system accessed and used directly by the facilities in a jurisdiction highlights the need to work directly with facility and field staff on a routine basis. The data in the resulting emission inventory have become more accurate with implementation of the system, which allows emitters to evaluate facility emissions estimates in comparison with reported process parameters, thereby adding a consistency check of the data.

Software is under development that integrates heterogeneous databases. Examples include applications in California to electric utility emission databases and to forest, controlled fire, wildfire, and smoke databases. User-friendly software based on geographic information system databases has also been developed. One example application is for the spatial location of emissions in the Juarez, Mexico–El Paso, Texas, area.

An initial national emission inventory is being prepared for Mexico. The inventory development follows guidelines from U.S. experience[2] and uses existing Mexican data, with gaps being filled by special studies. For example, versions of MOBILE6 and NONROAD models have been developed with data specific to Mexico. Data exchange protocols are being integrated to allow the Mexican data to be shared with interested parties in the United States and Canada.

Concluding Messages

The workshop concluded with a plenary address, a closing discussion by a tri-national panel, and concluding reports summarizing the technical presentations.

Peter Tsirigotis of the EPA's Emission Monitoring and Analysis Division recognized the pressing demands made on those charged with preparing emission inventories, which leave little time for innovative efforts to improve on existing approaches and methodologies. Within this context, however, he noted the importance of thinking "outside of the box," echoing the charge to the attendees made at the workshop's initial session. In so doing, he quoted the apt metaphor, "keep your nose to the grindstone and your eyes to the hills," and commended the attendees for their efforts in furthering this important cause. Tsirigotis also emphasized the critical importance of reducing and quantifying uncertainties in emission inventory development. He noted that substantial delays in U.S. acid-rain action resulted from disputed inventories during the late 1970s and early 1980s and that, in contrast, currently available emission inventories have enabled the proposal of numerous regulatory and legislative initiatives. In the future, further development along the lines of this workshop can help by doing the following:

- Providing more quantitative understanding of the relationships between management actions and their consequences;
- Indicating the implementation challenges of new approaches;
- Identifying windows of opportunity; and
- Focusing on issues of current concern.

In discussions after Tsirigotis' presentation, attendees noted that several governmental agencies, in addition to those charged with pollution management, require valid emission inventory data. Examples include U.S. and Canadian pollution forecasting programs and climate change research programs conducted worldwide. It was also noted that the workshop's discussions were closely related to the flow of information in air quality management (Figure 1), which illustrates the context of policy decisions after an information cascade that begins with

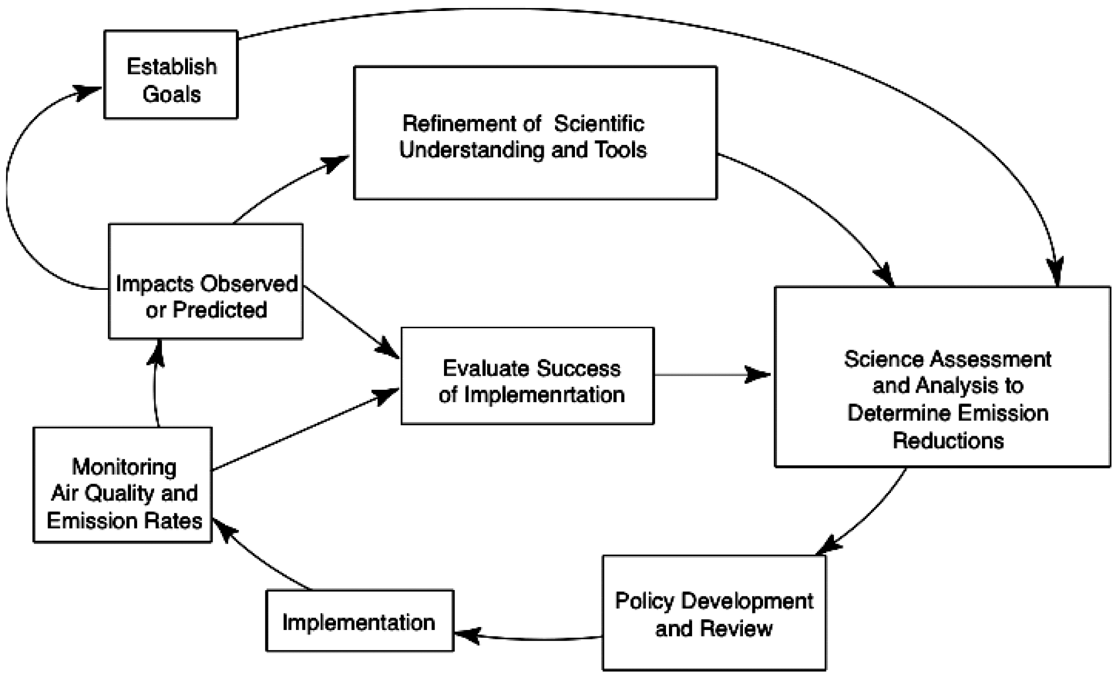

Figure 1. Flow of information in the development of air quality management policy (after NARSTO[5]).

emission and air quality measurement. Obviously, any uncertainties in emission estimates will propagate through this total sequence. Thus, process integrity is strongly related to individual reliability of each of the steps, including the emission characterization component. Although this seems self-evident, it is useful to keep in mind this composite cyclical structure, and its central dependence on process emissions.

The tri-national closing panel included Marc Deslauriers of Environment Canada, Phil Lorang of the EPA, and Leonora Rojas of Mexico's Instituto Nacional de Ecología. Group discussions interwoven with these speakers' presentations focused on differences and similarities in national approaches and future opportunities to work together on common goals. Numerous challenges and difficulties were noted, including speciation requirements for PM and VOC, dealing with process startup and upset emissions, fugitive emissions, and the temporal variability of emission sources in general.

Several attendees noted the need for developing a consistency or uniform methodology of information-gathering and -reporting practices. In this context, Canada and the United States are somewhat more aligned than Mexico, owing to the longer history of cooperation between the two northern countries. On the other hand, Mexico, with its recently developed National Emission Inventory, is moving ahead quickly. Dr. Rojas noted that Mexico would realize their national inventory by taking as much advantage as possible of U.S. and Canadian products, building a strong national capacity, and maximizing communications with external communities. Several emerging techniques, especially satellite methodologies, encourage the "harmonization" of national information because single observing platforms view areas of all three countries.

Specific Recommendations

Common themes from the six technical sessions again emphasized the importance of determining, quantifying, and reporting uncertainties in emission inventories, using top-down/bottom-up approaches systematically to develop these uncertainty estimates, and developing and applying new technology to these ends.

Source and Flux Measurements. Several techniques are currently being applied on limited scales for (mainly) bottom-up emission measurement. To further the application of these techniques, ad hoc consortia should be formed to facilitate information exchange with the policy and emission inventory development communities and to develop technological and programmatic acceptance of these emerging technologies. Additional observations and recommendations included the following:

- Advanced spectrophotometric techniques should be applied much more extensively for emission measurement in a variety of areas. Although not all these methods are new, the value of their ability to perform reality checks of calculated emissions is of increasing significance as emission inventories are consolidated and accuracy requirements are heightened.
- Communication between technical groups in the diversity of applications discussed in this session

- should be enhanced, perhaps by designating individuals to facilitate and coordinate objectives and efforts.

Mobile Source Characterization. Representatives stressed that this category includes a diversity of topics and new developments. Although many of these techniques are in practice to varying degrees, their applicability will continue to increase with continued advances in analytical instrumentation. Support should be given to improving PEMS and PAMS, remote sensing devices, and instruments capable of specialized measurements in roadside, tunnel, and chase studies. The forthcoming MOVES mobile source emission model has the potential of being a major step forward in emission inventory modeling. It will require considerable effort to acquire and validate new data, especially for nonroad sources.

Other observations and recommendations with respect to mobile source characterization were as follows:

- Paved and unpaved road dust models must be improved to account for real-world issues, including particle lifetimes in the atmosphere as a function of particle size and meteorology.
- Nonroad activity and emission data are extremely limited. Because diesels are important non-road sources, PM capability should be added to PEMS before major studies are performed.
- More work needs to be done measuring tire and brake-wear $PM_{2.5}$ emissions, because they are estimated to be greater in magnitude than gasoline LDV exhaust $PM_{2.5}$ emissions.
- There remains a need to be able to distinguish between gasoline and diesel PM emissions in ambient studies.
- Current activity data typically assume diesel vehicle miles traveled is a constant fraction of the LDV miles traveled. This assumption probably introduces major errors to the motor vehicle inventory and should be avoided.

Ground, Aircraft, and Satellite Observations. Both refinements of existing technologies and innovative conceptualization of new methodologies are equally important. Several types of satellite-based, as well as aircraft- and ground-based, observations are currently being applied for top-down emission inventory analysis, thereby helping to validate bottom-up inventories and providing associated uncertainty estimates. The potential for such applications is far greater than the level realized to date. In the case of satellite technology, the emission measurement community should communicate their needs directly to the satellite development community to encourage design and deployment of observing packages that are optimally useful for emission characterization.

The use of ambient concentration or atmospheric column burden data, supplemented with meteorological data and air quality models, offers major opportunities for verification of emission inventories, not only in North America but also across the Northern hemisphere. The mass balance and concentration ratio methods for gases and PM provide quantitative knowledge about emissions that complement conventional methods. These techniques can be greatly improved with the fast-response instrumentation now becoming available for ambient measurements.

Despite the promise of new techniques for verifying emission estimates, significant limitations in the application of conventional ambient concentration measurements and remote sensing data should be taken into account. In addition to sampling and measurement errors, many methods for bottom-up emission inventory reconciliation involve only a snapshot comparison in space and time. Inherently, the top-down methods have a basis that is inconsistent with emission estimates based on long-term averages. Furthermore, questions remain concerning the generalization of apparent inventory flaws based on the experience in specific cities such as Los Angeles, Mexico City, and Houston. Another question is whether or not the verification of conventional inventories can be performed through ambient measurements in a timely way so that corrections can be communicated readily to the regulatory community.

Additional observations and recommendations with respect to ground, satellite, and aircraft observations included the following:

- Extended analyses should be performed to determine whether the ambient concentration methods would provide an alternative basis for constructing or augmenting time-variable emissions inventories.
- Remote sensing methods show promise for certain qualitative and quantitative applications. To enable their widespread use in source characterization, even on continental and global scales, their uncertainties should be quantitatively defined, spatial resolution (vertical and horizontal) improved, and ways found to more closely link the column burdens to ambient concentrations at or near the earth's surface.

Air Quality and Receptor Modeling. Discussions focused on several model-based techniques for top-down emission analysis. Some of these, such as the receptor modeling and sensitivity/uncertainty analysis using deterministic models, have been applied in various forms for several years, but are evolving on a more-or-less continuous basis. Inverse modeling, on the other hand, is a technique relatively new to atmospheric sciences, although it has experienced considerable application in other fields such as groundwater hydrology. Viewed simply as executing a deterministic model backward to calculate sources on the basis of ambient concentrations, inverse modeling can be expected to receive increasing application during future years.

Modeling applications of the types discussed in this session tend to be complex, and several comments in the plenary session addressed the danger of "over-interpreting" model results to derive erroneous conclusions. Although these approaches are potentially powerful, informed interpretation is mandatory to ensure meaningful results. Additional observations and recommendations with respect to air quality and receptor modeling include the following:

- Formalized procedures should be developed to allow the results of receptor and inverse modeling tools to validate or correct estimates stated in conventional emission inventories when monitoring and modeling studies demonstrate inventories to be inaccurate. A mechanism should be established to allow procedures to be agreed upon to address circumstances (routine or special cases) for inventory correction. Establishment of a process to report a reliable inventory must address institutional impediments to the procedures.
- The strengths, weaknesses, and relative uncertainty expressed as ranges or probability of emission data created by emission estimation and modeling tools needs to be addressed as metadata in emission inventories to provide a more realistic representation of emissions. Because of the regulatory preference for quantification in terms of single numbers, it will be necessary to have institutional agreements for probabilistic representation.
- Inventory improvement tools and their resource needs should be integrated into the inventory building and validation process, rather than applied only after the fact.
- High-resolution temporal data (both emission inventory and ambient measurements) are needed for all source categories to enable the power of new emission modeling and evaluation techniques.
- Complementary uses of different methods should be used to perform cross-validation of emission inventories.
- Receptor and grid-based air quality models should be used jointly to better relate emissions with ambient concentrations.
- More collaboration is needed between the areas of ambient measurements, air quality modeling, and emission inventory development. Planning and scheduling of these activities should be conducted jointly.

Emission Modeling. Spatial and temporal variability in emissions and in emission modeling are important. Numerous challenges exist in this area (e.g., dealing with VOC and PM speciation, biogenic and fire emissions, and incorporating uncertainty and variability estimates in emission model output), but more recent measurement developments (e.g., continuous emission monitoring data) allow improvements as well. Observations and recommendations with respect to emission modeling include the following:

- Resources are limited and must be used efficiently. Therefore, efforts should be concentrated on the portions of the inventory that have both large emissions and large uncertainty (such as agricultural NH_3 and HDV off-road NO_x).
- In addition to improving current estimates, it is important to fill gaps in the inventory that may be important for new uses (e.g., PM modeling needs, data on fires, and different biogenic species).
- Effort should be made to obtain consistency in international, national, and regional inventories. This need reflects the ever-increasing global nature of air pollution control and therefore global inventories. This need is also linked to optimizing resources, because improvements made for regional studies could improve national inventories if they are included on a timelier basis.
- Consistent source identifiers should be applied to minimize double counting and facilitate cross-referencing. Simple methods such as using fixed identifiers for point sources in multiple databases, providing country codes, and sharing methods and results of updates are needed.
- The importance of spatial and temporal variability must be recognized. For example, in TexAQS 2000, biogenic VOCs represented about half of the VOC inventory, yet because of their spatial distribution they had little impact on peak concentrations in the urban area. Industrial emissions are often treated as constant, but highly variable industrial emissions were found to

contribute significantly to episodic peak concentrations.

- Increased use of probability distributions in emission estimates to better represent source variability has promise in helping to understand high pollution episodes. Tools are needed to characterize emissions variability (e.g., CEM data from electric generating units, industrial sources, and in-use motor vehicles) and incorporate the variability into emission and air quality models. Faster models are needed to run a multitude of scenarios reflecting emission variability. Detailed grid-based models are too expensive to provide the multitude of runs necessary to evaluate the impacts of emission variability on predicted concentrations, but the use of simpler models linked to baseline grid modeling can be useful.
- Open community-based emission models and processors are needed. They should be developed by users for users, with the codes open and available for users to make improvements and additions.
- Data sharing and data transparency are critical (e.g., consistency of land-cover data used in meteorological models that transfer into emissions models).
- Data-exchange protocols should be employed to help share data and build transparency. These protocols are intended to define common formats and mechanisms for sharing model-related data developed under different studies. They go beyond and build on the national emission inventory input format developed by the EPA to provide greater commonality.
- Quality assurance mechanisms should be built into the emission modeling and archiving processes (e.g., allowing comparisons with ambient measurements and spatial surrogate data). When emission inventory data are used in modeling studies, it is important to apply alternative techniques for evaluating the inventory in the context of model performance.
- Process-based models should be applied to help characterize and constrain emission estimates (e.g., NH_3 from concentrated animal feeding operations is limited by the nitrogen input; BVOC models rely on vegetative cover and meteorological data). Information on factors/activities/processes driving NH_3 emissions will be needed to inform emission modeling to accurately represent geographic, seasonal, and daily variability in ammonia emissions.
- Mechanisms should be developed to handle future projections that appropriately recognize variability in temporal and spatial factors as well as emission rates (importance may differ between Canada, Mexico, and the United States). In rapidly developing regions, it may be more important to reflect changes in land use and industrialization in future emission estimates.
- Communication with climate change projects to help generate ideas for inventory improvements is important.

Uncertainty and Data Management. Within the context of data management, new formalized procedures are needed to provide uniform approaches to top-down/bottom-up emission inventory development and data archival. In parallel, it is also essential to report uncertainties associated with emission inventory estimates. The TexAQS 2000 experience was cited as a prime example of this importance. Other observations and recommendations with respect to uncertainty and data management include the following:

- Emission inventory performance evaluations have reached the point that selected approaches can be formalized by incorporating them into guidance to states/provincial agencies and developers of emission inventories. Several appropriate approaches should be further refined and so codified.
- Once a model-generated emission inventory has been evaluated, the evaluation approaches should provide methods by which the emission estimates can be refined and rendered more accurate. Adjustment procedures should be defined to accomplish these improvements, and codified in the form of guidance. A means to trace the refinements back to each point source and make appropriate "ground truth" adjustments should be developed.
- The above recommendations should be sent to federal and state/provincial agencies for inclusion in their official guidance. The approach should be similar to that followed with chemical transport model performance evaluation.
- Evaluation of emission inventories with monitoring data is limited by the design of the ambient monitoring network and the airborne monitoring strategy. Increases in monitoring complexity and sophistication are obviously reflected in increased costs. When future field studies are designed, significant resources should be devoted to the collection of data for emission inventory validation and assessment. In addition, significant resources should be devoted to performance

evaluation of the emission inventories associated with the field study.

- Existing ambient monitoring networks should be evaluated and modifications should be considered so that the data collected are "optimally" useful for emission inventory performance evaluation as well as other measurement objectives.
- New state-of-the-science software that integrates data from various sources should be used as soon as possible to develop emission inventories. The results should be user friendly for agencies and facilities, comprehensive, integrated across and within the agencies, possess increased accuracy, and provide readily accessed data for reports.
- The EPA should support and establish a software development/refinement center that can be used by states and other users to selectively take modules and routines that can be universally adopted, adapted, and refined by individual users and included in their jurisdiction's own software to meet their individual needs.
- Data management systems and emission inventory development tools should be harmonized between Canada, Mexico, and the United States.

CONCLUSIONS

The takeaway messages from the workshop on the overarching science and policy questions are discussed below.

Policy Questions

What policy decisions will be made over the next several years based on current emission inventories?

Many applications for emission inventory applications across the United States, Canada, and Mexico are expected in the next few years. These include air quality forecasting, risk assessments, control strategy development, cap and trade programs, economic incentive programs, new source review, global climate, international transport, accountability, and assessments. With the first national emission inventory becoming available in Mexico, this country is especially poised to explore new applications.

What is the vulnerability of current approaches affecting these decisions?

Significant vulnerabilities exist within current emission inventory programs, which may limit their application or hamper their effectiveness. Most of these limitations are associated in some manner with the overarching challenges of data quality, representativeness, comprehensiveness, timeliness, and cost. However, significant capabilities are also apparent in the emission inventory programs, and improvements are steadily, if slowly, occurring. In many cases, emission inventories have supported major regulatory programs and have withstood legal challenges. Nevertheless, to position emission inventories to address program expectations in the future, improvements to current capabilities are expected to be necessary.

What new and innovative techniques can make a difference to these decision processes?

The workshop verified that science and technology advances are contributing new tools and techniques, which could and should be employed. These encompass new and innovative tools and techniques such as satellites, aircraft, and other remote sensing techniques, measurement and monitoring instrumentation, and protocols including continuous emission monitoring systems, computer software and hardware, including the Internet, and geographic information system capabilities, analyses, and application methods, including emission and air quality modeling, inverse modeling, and source receptor analyses.

Science Questions

Do newer and more innovative techniques exist on the "technical horizon"?

New techniques are available for near-term as well as long-term enhancements; however, miraculous revolutionary changes are not evident. Most of the methods discussed in the workshop were not new in the sense that essentially all of the methods have been used before. However, there are new techniques for established methods that are emerging and application of refinements to established methodologies.

What are they, what is their nature, and what are their limitations?

Improved measurement and analytical techniques are available across the spectrum of the emission inventory program. Innovative techniques that are emerging involve methods to establish inventory uncertainty using ambient data and specialized emissions studies, particularly for highly variable emissions from chemical plants, cooling towers, flares, and in-use motor vehicles. They have been improved substantially by new sampling techniques and fast-response instrumentation. They are inherently limited by resources to study a few cases for short periods of time, which precludes a robust statistical approach for long-term averaging and probability analyses. Remote sensing techniques, including satellite imagery, give qualitative knowledge about the large-scale emission patterns of fires and industrial or urban plumes. Recent advances in quantifying large-scale emissions of NO_2 and, indirectly, BVOC emissions have been developed from satellite data. All of the methods described can be adopted in principle for evaluation of conventional inventories. However, many of these tools and techniques require more research and development, additional resources, and increased attention to detail, which will limit their use by programs with resource constraints.

How can these methods best be combined with conventional methods, as well as with other more modern techniques?

Evolution of emission inventory programs is integrating many improvements on an ongoing basis. The methods can be combined with conventional methods by intercomparison of the results expected from emission models with air quality modeling, or consistency checks with ambient data using, for example, ratios of concentrations representative of specific sources. The results to date still rely heavily on the conventional approach to establishing emission factors, activity patterns, and emission control estimates; there has not appeared any kind of new "emission paradigm" for estimations. There is a need, however, to formalize intercomparison methods for establishing the reliability of international consistency of inventories for many applications.

Next Steps

The identification of potential new tools for the development and improvement of emission inventories begun at the workshop will be continued; an assessment document, following in the tradition of the O_3 and PM assessments prepared under NARSTO sponsorship,[4,5] is under development. The Emission Inventory Assessment is intended to provide a roadmap for improving inventories, promoting efficient and effective use of inventories, and guiding future applications of emission inventories in air quality management.

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