CHAPTER 6

INNOVATIVE TECHNOLOGIES AND APPLICATIONS

Chapter 5 described strengths and weaknesses of current emission inventories. Addressing the weaknesses of inventories and of the methodologies used in developing them has been an important technical activity for at least two decades. It has led to major advances in measurement and observational capabilities, as well as improved modeling and interpretative methods. In addition to these advances, modern information-management techniques offer the opportunity for greatly improved inventory accessibility and transparency. Chapter 1 noted that contemporary research offers new and evolving techniques that are potentially applicable for developing and/or verifying emission inventories, and that these methodologies should be considered for broad application in the future. This chapter provides a brief overview of these techniques.

Three general aspects of these techniques and methodologies are noteworthy: First many, if not all, of the methods discussed here are not really “new;” indeed, most have been in existence in one form or another for a number of years. Receptor modeling and plume measurement by ground-based optical remote sensors, for example, have been evolving for some time. Other techniques, including satellite remote sensing and inverse modeling, are relatively new to air pollution analysis, though the latter has experienced extensive application in other fields. Key points to note in this respect are that the selected methodologies are evolving in application, leading to new approaches for emission inventory analysis. It is important that workers in the emission inventory field monitor these developments, and take advantage of their capabilities to address weaknesses in the historical approaches to inventory development.

The second noteworthy aspect is the fact that the potential for innovative application often lies more in a combination or integration of two or more of these technologies, rather than in isolated deployment of a single technique. Creative combinations of aircraft remote-sensing measurements, ambient concentration observations, and demographic or source data from a well-designed field campaign with an inverse-modeling analysis, for example, can reveal substantial information regarding specific emission sources.

Third, many of the technologies discussed in this chapter were not developed specifically for emission inventory application, but subsequently have proven useful for emission analysis. The emission inventory community needs a process that is more direct and focused than this largely happenstance approach. Accordingly, an active dialog between emission inventory developers and scientists involved in adopting measurement and interpretive methodologies is vital. Inventory developers need to increase their awareness of technological developments related to their field; but more importantly, they also should think creatively about their future requirements and communicate these requirements to their counterparts so that future development occurs less by chance and more by intentional design. A dialog between emission inventory scientists and those developing sensors for satellites is a prime example of such an interaction. The sections of this chapter discuss examples of these evolving methodologies as a starting point for developing better inventory-development techniques.
6.1 OBSERVATION AND MEASUREMENT METHODS

Advanced measurement capabilities can address many of the weaknesses in current emission inventories described in Chapter 5. Important applications for new observational opportunities include:

• Seeking complementary and efficient approaches to conventional stack and effluent testing, including compositional data
• Expanding the use of a variety of ambient concentration data to cross-check inventory estimates and provide quality-control options
• Establishing a linkage with “real world” sources, including vehicle fleets, compared with idealized emission conditions
• Expanding spatial and temporal data for mobile and area sources to improve emission models
• Providing direct measurements of biogenic and fugitive emissions for incorporation into emission models
• Providing a basis for determining whether long-term, estimated emission trends are consistent with ambient concentrations.

This section describes a number of evolving observational and measurement techniques having emission inventory applications, summarizing them in sufficient detail to give the reader a basic idea of their measurement principles, their future potential, and their limitations. As is readily apparent in the following sections, some of these methods are much more complex than others, and require more extensive descriptive detail to convey their features. These differences are reflected by the selected lengths of the individual descriptions presented below, which are arranged in descending order of complexity and attendant descriptive detail.

6.1.1 Remote Sensing

Remote-sensing techniques (cf. Stephens, 1994; Finlayson-Pitts and Pitts, 2000) fall into a number of classes depending on the type of sensing signal applied (e.g., electromagnetic or sonic), electromagnetic wavelength range (ultraviolet, visible, infrared, or microwave), whether the method in question observes spectrally disperse or broadband radiation, whether it has ranging capabilities, and the type of signal detected (e.g., absorption, light scattering, fluorescence, . . .). Several systems within these categories have been deployed to determine emissions from both stationary and mobile sources, using surface, aircraft-based, and satellite-mounted sensors.

In general, remote-sensing chemical measurements sense path-integrated loadings, or at best (in the case of ranging measurements), ambient concentrations. Deriving actual emission rates from these data requires either knowledge of concentration distributions along a sight path and a direct or indirect indication of flow velocity, or else the use of index species (such as CO$_2$) in conjunction with process-stoichiometric calculations. In some cases, such as satellite or aircraft surveillance, an air quality model is required to infer emission rates. Although these deficiencies may be resolved during future years by application of advanced techniques such as tomography for path-measurement reconstruction and laser, microwave, and/or sonic anemometry for flow measurement, they should be borne in mind as significant issues in the present context.

Electromagnetic radiation remote-sensing applications fall into several categories, the most important of which are summarized as follows:

Absorption Spectroscopy. Open-path, absorption-spectroscopy techniques produce path-integrated measurements, and monitor attenuation of a light beam as it transects the sampled atmosphere. As

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1 Light absorption and emission by molecules and atoms occurs as a consequence of energy transitions between quantum states, and thus individual pollutants have characteristic spectroscopic signatures, which can be used for their identification and measurement. Individual “signatures” often dictate which portion of the electromagnetic spectrum is most appropriate for measuring a specific pollutant. Because of its comparatively high photon energy, the ultraviolet/visible portion of the spectrum is associated mainly with transitions between electronic energy levels. Lower-energy infrared radiation is associated with energy transitions between various molecular vibrational and rotational energy states; microwave radiation, which has even lower photon energy, interacts mainly with rotational transitions. These features usually dictate the choice of one technique versus another for observation of a specific pollutant. One should note that besides these molecular-level interactions, radiation can be scattered physically through interactions with atmospheric density fluctuations, airborne particles and precipitation elements.
such they require a sensor viewing a remotely located radiation source, or else a source co-located with its sensor, viewing a remote reflector.

Variants of absorption spectroscopy include non-dispersive infrared (NDIR) techniques, which measure the attenuation of a broadband radiation source by the sampled medium. This approach is applicable to pollutants such as CO, whose broadband absorption spectrum dominates those of other gases in specific spectral regions. Dispersive techniques, in contrast, typically deploy a continuous light source and a detector incorporating a diffraction grating to disperse the incoming radiation according to wavelength, or else a laser light-source which may be tunable over a limited wavelength range. Variants of these methods include Fourier transform infrared (FTIR) methods, which use an interferometric sensing approach to generate a Fourier transform of the spectral signal, and thus can monitor the entire spectral range essentially simultaneously. Because of its spectrally resolved approach, FTIR can measure emissions of a large variety of compounds. Yokelson et al. (1997), for example, applied FTIR to measure formaldehyde, ethanol, acetic acid, ethene, propane, propanol, HCN, CO, CO$_2$, CH$_4$, and NH$_3$ emissions from combustion processes. Another technique, differential optical absorption spectroscopy (DOAS), applies a broadband light source, disperses the incoming signal into a resolved spectrum, and applies an interpolation process to estimate the spectral background, which is subtracted from the total spectral signal to obtain the pollutant-induced component. Both FTIR and DOAS can operate in a passive mode, using natural light sources such as the sun or moon. Positioning requirements, however, generally dictate the use of active techniques in the case of pollutant emission analysis.

Fluorescence Spectroscopy and Raman Spectroscopy. Fluorescence-based methods transmit a light beam through the sampled atmosphere to induce electronic excitation of pollutant molecules, which emit radiation when transitioning back to their ground states. The emitted radiation is monitored by a sensing device, providing a measure of pollutant concentration or pathway loading. Excitation of specific molecules, as well as their resulting fluorescence, is wavelength-specific; thus single- or dual-wavelength lasers, selected for the specific pollutant of interest, are applied most often for this purpose. In their simplest form, open-path fluorescence-based methods produce path-integrated results, but can incorporate ranging when applied in conjunction with lidar systems (see below). Raman spectroscopy operates in a manner somewhat similar to standard fluorescence spectroscopy. Here, however, the frequency of the incident light beam is shifted to a (usually) lower value by extraction of a portion of the photon energy through interaction with the sampled medium.

Light Detection and Ranging (lidar). Lidar is based on projecting a coherent light beam through the sampled air volume and monitoring the return signal, which results from light scattering by the target pollutant material. In contrast to the methods described above, lidar has a range-gating capability, which allows generation of pollutant-profile information at relatively fine intervals (down to about 3 m). Single-wavelength lidars have been applied since the 1960s to remotely sense PM concentrations, and more elaborate, multi-wavelength lidars (e.g., differential absorption lidar) have been applied increasingly to measure spatial distributions of trace gases. A Raman spectroscopy lidar variant also has been applied during recent years.

All of the techniques described above must address issues of sensitivity and specificity, which are highly species- and technique-dependent but of some concern in practically all cases. The associated equipment tends to be expensive and in many cases requires highly experienced operators. Moreover, and as discussed above, these detection methods generally depend on ancillary effluent flow measurements or on inferred flows based on modeling or stoichiometric ratioint techniques to determine actual emission rates.

The following sections, which discuss remote...
sensing from satellite-, aircraft-, and surface-based platforms, provide some examples of these issues and their resolution.

6.1.1.1 Satellite Remote-Sensing Applications

Measurement and Interpretive Bases

Although satellite measurements of surface features such as vegetative ground cover, surface temperature, and ocean activity are of some interest in an emission inventory context, actual pollution loadings are of more direct utility and provide the most difficult challenges. Consequently these applications will be emphasized here. Up to now satellite observations of trace gases and PM in the troposphere have been confined largely to passive, downward-looking, spectrally resolved techniques. These techniques observe energy emanating from the surface or the atmosphere and derive concentrations and/or column densities from the amplitudes of spectral lines at specific wavelengths associated with the pollutant molecules of interest. Currently operational (and most planned) measurements are made from low Earth orbit, providing a swath of data during each orbit with individual measurements on spatial scales of tens to hundreds of kilometers (cf. Figure 6.1).

Perhaps the best known applications of satellite measurements for emission estimation involve the imaging of the planet surface to identify the location and intensity of smoke sources, ship plume tracks, major industrial plumes, and incidents of dust storms or regional haze. These visual images have aided in identifying potential source areas of interest for smoke and haze emissions and transport. However, they are extremely limited in their ability to provide quantitative data. Important progress has been made recently in using the satellite data to infer pollutant column concentrations or densities, and continental-scale emissions.

Although satellite applications for determining trace-gas and PM emissions must deal with numerous challenges, important advances have been made in identifying and quantifying these emissions. Figure 6.1 demonstrates the utility of satellite data in identifying and characterizing NOx emissions from isolated, individual cities such as Jeddah, Mecca, Medina, and Kuwait City.

Figure 6.1. Tropospheric NO\textsubscript{2} column densities in the Red Sea/Arabian Gulf area derived from SCIAMACHY data for September 2002 (after Richter, in Borrell, et al. 2004). Emissions of NO\textsubscript{x} from isolated, individual cities such as Jeddah, Mecca, Medina, and Kuwait City are easily identified, based on the conventional assumption that NO\textsubscript{2} is a surrogate for NO and NO\textsubscript{2}.

\footnote{LANDSAT data have been used on a regional scale, for example, to support evaluation of changes in biogenic emissions in central California (Tanner et al., 1992).}
technical challenges, their ability to cover large, typically global, spatial domains provides a major advantage not shared by other approaches. This global coverage encourages the application of European, as well as North American satellite observations for emission evaluations over large areas of the NARSTO domain. The European research community is making considerable progress in this field (Borrell et al., 2004), and indeed data from European satellites are being applied currently for interpretation of North American emissions (e.g., Palmer et al., 2003a).

Many of these observations are based on UV or visible light reflected or backscattered from the surface or from clouds, and derive vertical-location estimates from the spectral-line widths, which result largely from pressure broadening. These techniques are available only during daylight hours and provide no information at night. Other passive techniques operate in the infrared portion of the spectrum, which is available continuously throughout the day and night. Usually, vertical location is derived from infrared (IR) measurements based on the variation of radiative properties with atmospheric temperature, thus vertical temperature structure is an important data-processing consideration. Satellite PM observations have been demonstrated using lidar, an active technique which can provide detailed vertical information under most conditions. Lidars produce sparser horizontal data sets than passive systems because greater instrument energy is required.

Typically, the inference of emission rates from satellite measurements is derived in two general steps: (1) retrieval of lower-atmosphere pollution concentrations from the raw satellite data and (2) estimation of emissions on the basis of this near-surface information, usually in conjunction with ancillary data and interpretive calculations. Substantial processing is required in the retrieval stage to convert raw satellite data into useful products, such as concentration patterns, column densities, and optical depths, and the retrieval algorithms applied for this purpose tend to be complex. Estimation of emission rates from retrieved satellite products also requires substantial processing and interpretive effort.

The following subsection gives several examples of different approaches in this general area.

Satellite-based measurement of tropospheric pollutants presents several technical challenges. These include 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. Satellite measurements beneath cloud cover are virtually impossible with present technology. Moreover – with the exception of lidar – attempts to resolve measurements vertically typically depend on interpretation of secondary effects such as pressure broadening and temperature influences, thus limiting the resolution of vertical structure. In fact, satellite-based trace-gas measurements have not yet resolved even two separate layers in the troposphere. Further, satellite instrument sensitivity is often a strong function of altitude. Thus, each instrument has its own characteristic averaging kernel for each species, which defines the altitude dependence of its sensitivity. As a result, retrieval of integrated concentration profiles requires information regarding the species’ vertical distribution, and leaves a significant uncertainty regarding the derived quantities. These features combine to result in highly species-specific retrieval algorithms, and limit the number of tropospheric pollutants that can be observed reliably; however, this limited set includes important pollutants such as PM, and a number of key secondary-pollutant precursors.

Table 6.1 summarizes existing, planned, and feasible satellite-based tropospheric trace-gas and PM measurements by the U.S. National Aeronautics and Space Agency (NASA) and the European Space Agency (ESA). Future measurement systems that are feasible with current sensor technology include geostationary trace-gas measurements, which can provide essentially continuous coverage (many observations per day over the same location) at high horizontal resolution. As indicated by Table 6.1, satellites currently in use revisit portions of the globe only periodically, usually at the same time of day, and thus provide less temporal coverage than is usually desired. This issue may be resolved during

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Table 6.1 is not visible in the text provided.

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More comprehensive information on North American and European satellite programs is available on the NASA and European Space Agency (ESA) websites: www.earth.nasa.gov/ese_missions/satellites.html and envisat.esa.int.
Table 6.1. Selected Past, Current, and Future Remote Sensing Instruments used to Determine Amount and Distribution of Constituents in the Troposphere.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Name</th>
<th>Vertical extent of measurement</th>
<th>Horizontal resolution, domain</th>
<th>Temporal revisit</th>
<th>Target Constituent/Property for Air Quality</th>
<th>Platform</th>
</tr>
</thead>
<tbody>
<tr>
<td>GOME</td>
<td>Global Ozone Monitoring Experiment</td>
<td>Troposphere and Stratosphere</td>
<td>40 x 40 km², 40 x 320 km² swath</td>
<td>Once every 3 days</td>
<td>Tropospheric columns for O₃, NO₂, BrO, SO₂, HCHO, clouds and aerosols</td>
<td>ESA-ERS-2 (1995-present)</td>
</tr>
<tr>
<td>MODIS</td>
<td>Moderate Resolution Imaging Spectroradiometer</td>
<td>Surface to space</td>
<td>0.25 - 1 km, 2330 km wide swath</td>
<td>Once 1-2 days</td>
<td>Aerosol column optical thickness, aerosol type (sulfate, biomass burning) over land</td>
<td>NASA Terra (1999) NASA Aqua (2002)</td>
</tr>
<tr>
<td>MISR</td>
<td>Multi-angle Imaging SpectroRadiometer</td>
<td>Surface to space</td>
<td>0.275 - 1.1 km, 141 x 563 km² swath</td>
<td>Once every 9 days</td>
<td>Aerosol properties (angular radiance dependence)</td>
<td>NASA Terra (1999)</td>
</tr>
<tr>
<td>MOPITT</td>
<td>Measurement of Pollution in the Troposphere</td>
<td>Tropospheric columns, layers</td>
<td>22 x 22 km², 22 x 640 km² swath</td>
<td>Once every 3 days</td>
<td>Total column of CO, CH₄; CO layers</td>
<td>NASA Terra (1999)</td>
</tr>
<tr>
<td>SCIA-MACHY</td>
<td>SCanning Imaging Absorption spectrometer for Atmospheric Chartography</td>
<td>Tropospheric columns</td>
<td>30 x 30 km 960 km swath</td>
<td>Once every 3 days</td>
<td>Tropospheric columns for O₃, NO₂, N₂O, CO, CH₄, SO₂, HCHO, clouds and aerosols</td>
<td>ESA Envisat (2002)</td>
</tr>
<tr>
<td>OMI</td>
<td>Ozone Monitoring Instrument</td>
<td>Stratospheric profiles, Tropospheric columns</td>
<td>48 x 48 km²</td>
<td>Once per day</td>
<td>Tropospheric columns for O₃, SO₂, HCHO, NO₂, and aerosol</td>
<td>EOS Aura (2004)</td>
</tr>
<tr>
<td>TES</td>
<td>Total Emission Spectrometer</td>
<td>Stratospheric profiles, Tropospheric layers</td>
<td>26 x 42 km²</td>
<td>~ once every 2 days</td>
<td>Tropospheric columns for O₃, NO₂, CO, SO₂, CH₄</td>
<td>EOS-Aura (2004)</td>
</tr>
</tbody>
</table>
Table 6.1. Concluded.

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Platform</th>
<th>Target Constituent/Property for Air Quality</th>
<th>Temporal revisit</th>
<th>Horizontal resolution, domain</th>
<th>Vertical extent of measurement</th>
<th>Future instruments scheduled to be launched</th>
</tr>
</thead>
<tbody>
<tr>
<td>CALIPSO</td>
<td>NASA CALIPSO (2005)</td>
<td>Aerosol column loading and radiative properties</td>
<td>Not operated continuously</td>
<td>STRatospheric profiles, Tropospheric profiles</td>
<td>0.3 x 0.3 km²</td>
<td>Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations</td>
</tr>
<tr>
<td>Geo-TRACE</td>
<td>NASA Earth probe</td>
<td>Tropospheric columns for O₃, NO₂, SO₂, H₂O, CH₄, HCHO, CH₄, CO, clouds and aerosols</td>
<td>Once per hour</td>
<td>4 x 4 km², 8000 x 4000 km² (entire North American continent)</td>
<td>25 x 25 km²</td>
<td>Geostationary SCanning Imaging Absorption spectrometer for Atmospheric Chartography</td>
</tr>
<tr>
<td>Geo-SCIAMACHY</td>
<td>Potential future ESA mission</td>
<td>Tropospheric columns for O₃, NO₂, SO₂, H₂O, CH₄, CO, clouds and aerosols</td>
<td>Twice per hour</td>
<td>Entire Earth disk, European view</td>
<td></td>
<td>Future instruments scheduled to be launched</td>
</tr>
</tbody>
</table>

Finally, satellite observations typically report data in terms of average concentrations, column densities, or derived quantities such as optical depth; thus, any inference of emissions from such products necessarily depends on the application of inverse modeling, or some other interpretive technique.

**Example Applications**

The objective of this section is to provide the reader with an initial appreciation of available satellite products, and to demonstrate these products’ potential applicability for emission assessment. Figure 6.1 provides a simple example for an initial approach to this objective. Here, observation swaths from ESA’s SCIAMACHY show tropospheric NO₂ column densities observed in the vicinities of some Middle Eastern cities. Because of these cities’ isolation from confounding sources, this image immediately suggests that local NOₓ emission rates could be inferred from these data in conjunction with observed winds and using chemical-transport model analysis to simulate NOₓ chemistry.

Figure 6.2 shows tropospheric column densities of selected pollutants obtained from a variety of North American satellite sensors (Neil, Fishman, and Szykman, 2003). Such data provide a more complicated (but also more typical) example of large-scale burden patterns. If these burdens are confined to the planetary boundary layer, for example, they can be used with an air quality model for semi-quantitative, large spatial-scale source allocation. These plots indicate the potential complexity of inferring emission rates from continental data, which reflect long-range transport and multitudes of individual sources. The following paragraphs present some examples of studies where inroads are being made in this area.

Given the measurement challenges noted above, emission assessments using satellite-derived products are currently at a relatively nascent, but evolving, state. Interpretive work has consisted largely of studies wherein satellite-based column loadings of a pollutant or its reaction product are observed and future years for both North America and Europe by the deployment of sensors on geostationary satellites, providing almost continuous temporal coverage (see Geo-TRACE and Geo-SCIAMACHY in Table 6.1).
Figure 6.2. Typical satellite-derived tropospheric column loadings of selected pollutants, which are confined mainly to the planetary boundary layer. In the NO₂, PM, and SO₂ displays bluer or more yellow hues qualitatively indicate regions with relatively low emission rates. In all displays the orange to red or darker hues qualitatively indicate higher column loadings, identified with areas of higher emission density. Data obtained from a variety of satellite sensors (Neil, Fishman and Szykman, 2003).
compared with model-derived column loadings based on an assumed emission inventory. Subsequently the model is executed repeatedly, adjusting the emission inventory until a match between the model- and satellite-derived values is achieved. For cases involving multiple sources this can be viewed as a rather broad-brush approach, providing coarse estimates of overall emission magnitudes. These estimates are perhaps most useful (a) for large-scale geographical regions, where little or no quantitative information exists, or (b) for estimating natural vs. anthropogenic emissions.

Examples of this approach include the work of Petron et al. (2004), who applied MOPITT CO data in conjunction with the NCAR Model for Ozone And Related chemical Tracers (MOZART) to infer CO emissions from western U.S. wildfi res occurring during August 2000. Similarly, Palmer, et al. (2003b) applied formaldehyde column data derived from GOME measurements to infer North American emissions of the biogenic formaldehyde precursor, isoprene, using the GEOS CHEM chemical-transport model as an interpretive tool. As can be noted in Figure 6.3, comparison of GOME data with GEOS CHEM simulations based on two existing isoprene emission inventories suggests significant biases in these inventories.

A third example of this approach is the work of Martin et al. (2003), who applied GOME-derived NO\(_2\) column data to reduce errors in the global NO\(_x\) emission inventory. In some respects model applications of this type can be considered an elementary form of inverse-modeling applications. More formal inverse-modeling approaches, described in Section 6.2.2, involve a substantially more detailed mathematical treatment, but offer the possibility of increased resolution of individual pollution sources. To date this more formal approach has been limited to a few special applications, such as global CO\(_2\) emissions (e.g., Kasibhatla et al., 2003), but one can expect extended application in the future.

The current evolutionary state of satellite measurements should be expected to expand in at least three general areas. The first of these involves the realization of more highly specific measurements with regard to chemical species, vertical and horizontal resolution, and temporal coverage, which will be provided by platforms and sensors currently planned or envisioned. Progressively higher-quality satellite-based measurements of O\(_3\), NO\(_2\), SO\(_2\), HCHO, CO and PM will be available over the next five years from the recently deployed ESA SCIAMACHY and NASA’s EOS Aura (http://eos-chem.gsfc.nasa.gov/project). Moreover the geostationary platforms noted in Table 6.1, if deployed, will substantially improve spatial/temporal coverage of the North American and European continents.

The second important development involves interpretation of quantitative emissions. This has a somewhat longer time horizon, and depends strongly on the ability of scientists working in the emission inventory field to envision new satellite-derived products desired for their specific purposes and to communicate these ideas to their counterpart scientists in the satellite community.

Finally, as demonstrated by the example applications described above, considerable development can be expected in inverse-modeling applications and other innovative interpretive techniques, often using satellite measurements in conjunction with surface-based measurements. With developments in these areas, satellite applications can be expected to attain significantly greater source-resolving power, which may be a complement to conventional estimation methods.

### 6.1.1.2 Aircraft Remote-Sensing Applications

Both surface- and aircraft-based remote-sensing applications to evaluate pollutant emissions have relied almost totally on open-path optical techniques, and share considerable commonality with sensors currently deployed on satellite platforms. In contrast to most satellite-based approaches, a majority of surface and aircraft-based sensors deployed for emission assessment observe radiation emitted by the measurement device, and thus are classified in the “active” category.

Aircraft remote sensing provides a useful complement to in-situ emission measurements from aircraft, which are described in Chapter 7. As with their in-situ counterparts, aircraft remote sensing is usually applied for determining pollutant fluxes through vertical planes encompassing the plumes being
observed. Typically, such measurements are made by DOAS (Melamed et al., 2002 and references therein), infrared spectroscopy (Stearns et al., 1986), or by lidar.

Remote sensing has several advantages in aircraft applications. Most importantly, these methods provide a measure of the vertical column of the concentration through the plume, thereby directly evaluating an integral of pollutant concentration over the vertical dimension. The evaluation of integrated plume concentration is then reduced to integrating the column measurement across the plume. The plume flux can be determined using the average wind speed [See Equation (7.1)]. A second advantage is that the column measurement is insensitive to variations in the height of the planetary boundary layer (PBL) and vertical inhomogeneities of concentrations. Finally, the required cross-sectional measurement is accomplished in a single aircraft transect that can be

Figure 6.3. Comparison of North American isoprene emissions derived from GOME formaldehyde data for July 1996, with those estimated by the GEIA and BEIS2 inventories. From Palmer et al. (2003a,b).
carried out above the PBL. It is thus more suitable for determining plume-wide fluxes from extended sources, such as urban areas. One disadvantage of the technique is that the species that can be measured are presently limited; plume fluxes have been reported only for DOAS measurements of NO₂ (which allows the calculation of corresponding NOₓ fluxes) and for SO₂. The reported precisions for these measurements are near plus or minus 30 percent. In intense biomass burning emission plumes, molar ratios of a variety of species have been determined (Worden et al., 1997).

As previously mentioned, current aircraft-deployed, remote-sensing applications for emission analysis are limited mainly to DOAS measurements of NOₓ and SO₂ fluxes and IR spectroscopy determinations of emission factors for species released in biomass burning. DOAS techniques could potentially be extended to some VOC species including formaldehyde, alkenes and aromatics. There is also potential for lidar instrumentation to be applied to measured primary PM emissions. It is possible to remotely measure wind speeds using Doppler lidar techniques; application of this instrumentation would be a useful advance. The limiting factor in these possible projections will be the ultimate signal-to-noise ratios that can be achieved.

6.1.1.3 Ground Based Remote Sensing Application

Stationary Sources

For discussion purposes it is convenient to subdivide surface-based remote sensing of emissions into two categories, depending on whether the source in question is mobile or stationary. Remote-sensing evaluations of stationary-source emissions have applied several of the optical methods summarized in the introduction to this section. While having significant potential, they are limited for stationary source applications because of detection limits, especially for toxic gases at very low effluent concentration. Often used more for pollutant concentration studies not directly related to emission inventory evaluation, these techniques are currently much more limited for stationary sources than their mobile source counterparts. North American applications, for large point sources at least, have been superseded by the reliance on standard reference methods and CEMS, which often provide the most straightforward and direct approaches to emission measurement. On the other hand, remote sensing is often attractive in situations (such, for example, as petroleum refineries) involving complex source configurations and/or detection of fugitive emissions. The availability of commercial, off-the-shelf equipment for this purpose is limited, although a few organizations – particularly in Europe – offer services applying remote sensing for emissions from sources such as refineries and feedlots (e.g., www.spectrasyne.ltd.uk; www.kassay.com/kfshome.htm; www.erg.com/services/environ_meas_source.htm).

Several examples of successful application of remote-sensing technology for stationary-source emission assessment can be cited. Apart from the previously cited application of FTIR by Yokelson et al. (1997) to determine combustion-process emissions, these include the work of Schröter et al. (2003) who applied lidar measurements of a power plant plume to remotely sense SO₂ concentration, and processed the results with co-located sound detection and ranging (sodar) flow measurements to estimate SO₂ emissions. Schäffer et al. (2004) applied simultaneous upwind and downwind DOAS measurements to determine emissions from automobile service stations and from tanker filling operations, using a small-scale dispersion model to back-calculate emissions. Galle et al. (2001) applied active FTIR to monitor CH₄ emissions from landfills and NH₃ emissions from agricultural manuring applications. They also applied passive FTIR (using the sun as a radiation source) to determine emissions from petroleum-processing complexes and harbor operations. Haus et al. (1998) applied FTIR analysis of CH₄, CO₂, CO, NO, and water to determine emission rates by natural-gas flares.

A survey of this subject indicates that remote-sensing technology has large future potential for evaluation of emissions from complex sources, such as refineries or chemical plants, which are not amenable to standard single-stack treatment. Reflecting this future potential, the U.S. EPA is currently supporting research and demonstration programs applying remote sensing to a variety of fugitive emission sources (www.epa.gov/ORD/NRMRL/scienceforum/thoma_e.htm), and a number of innovative techniques
such as the application of tomography to FTIR path measurements for determining spatial distributions (Hashmonay and Yost, 1999; Hashmonay et al., 1999) – are currently in a development stage. Although it has received little application to date, remote sensing of effluent velocity fields in conjunction with pollutant measurements has a large potential for emission quantification, and can be expected to expand considerably during future years.

Mobile Sources

As noted in the previous subsection, mobile-source remote sensing has received significantly more application for emission analysis than its stationary-source counterpart, and several commercial applications are currently in operation. The most important of these is cross-road sensing using electromagnetic radiation, which performs a series of light-absorption measurements intercepting exhaust plumes behind moving vehicles. The initial implementation of this technology involved NDIR measurements of CO and CO₂, with CO₂ serving as the internal plume tracer. Stoichiometric ratios of excess (above background) plume CO/excess plume CO₂ can be used to compute the fraction of CO in the exhaust at the vehicle’s tailpipe (Bishop et al., 1989; Stedman, 1989). The measured excess target pollutant/excess CO₂ ratio also can be used along with a combustion equation to provide a target pollutant emission index (g pollutant emitted/kg fuel consumed). This system has been extended to measure HC and NOₓ emissions using NDIR absorption, and, with greater selectivity and sensitivity, using UV spectroscopy (Guenther et al., 1995; Bishop and Stedman, 1996; Popp et al., 1999). Similar NDIR cross-road instruments were also successfully developed by General Motors Research Laboratories to monitor CO and HC exhaust emissions (Stephens and Cadle, 1991; Cadle and Stephens, 1994). Commercial instruments based on this technology and methods of exploiting advances in software and computer hardware for improved instrument control and signal processing are currently produced by Environmental Systems Products, Inc. (ESP, 2003a).

Advanced cross-road remote-sensing systems based on tunable IR laser differential absorption spectroscopy (TILDAS) have been developed and deployed by Zahniser and co-workers (Nelson et al., 1998; Jiménez et al., 1999; Jiménez et al., 2000a,b). These systems have the advantage of longer measurement path lengths, more easily manipulated light paths, and greater sensitivity for a range of individual exhaust species, allowing more pollutants to be quantified more specifically. TILDAS measurements of exhaust NO, NO₂, N₂O, and NH₃ have been reported and calculations show measurements of other species such as CO, H₂CO, CH₃OH, C₂H₄, and CH₂=CH-CH=CH₂ are feasible.

Dispersive IR spectroscopy also can be used to quantify exhaust emissions from moving vehicles, although cross-road path lengths and vehicle speeds may be restricted, especially compared to the TILDAS systems. Baum et al. (2000) have demonstrated a prototype onroad vehicle emission measurement system combining IR and UV spectrometers. This system can quantify exhaust CO, NO, NO₂, N₂O, HONO, NH₃ and as well as some light aldehydes, aromatics, and aliphatic hydrocarbons, and has been used to measure NH₃ emission distributions on a Los Angeles freeway onramp (Baum et al., 2001). Finally, instruments to measure exhaust PM are currently being developed and demonstrated (Moosmüller et al., 2003; ESP, 2003b). ESP’s latest commercial remote-sensing technology includes a UV spectral measurement to generate a smoke index (g particles emitted/kg fuel consumed).

Cross-road remote-sensing studies have been extremely valuable in characterizing onroad emissions for light duty gasoline-powered vehicles. A critical finding from these fleet-emission measurements indicates that a small fraction of the vehicles emits a large fraction of a given pollutant, demonstrating that emission factors do not follow a normal distribution. Ten to 20 percent of the vehicles tested account for 50 to 80 percent of the emissions. These high emitters constitute a large part of the inventory, which is not well reflected by average emission factors. Onroad tests show that inspection and maintenance (I/M) programs often do not identify the highest emitters.
This highly skewed distribution was recognized by Zhang et al. (1994) for CO and HC emissions, and is also reported for CO and HC by Stephens (1994). The same distribution also has been shown to characterize NO (Jiménez et al., 1999) and $N_2O$ emissions (Jiménez et al., 2000a). This fact has a large impact on the sample size of light-duty vehicles whose emissions must be evaluated to determine statistically valid inputs for mobile emission models. Referring to enforcement practice, other studies have qualified the measurement of specific gas components as independent measures guiding vehicle inspection and maintenance surveillance (e.g., Mazzoleni et al., 2004).

Relatively long time-series of remotely sensed emissions for light-duty vehicles are now available for a number of North American and European cities, allowing a determination of how well new cars meet regulatory standards and assessments of emission changes with vehicle age (Pokharel et al., 2003; Sjödin and Andréasson, 2000). Comparable data are available from enough cities around the world that the impacts of variations in maintenance practices can be recognized (Zhang et al., 1995). Data on vehicle emissions from several Mexican cities have been compared and contrasted with similar data from U.S. cities (Bishop et al., 1997). Studies like these are important to inform mobile-source emission models about expected temporal and geographic variations of mobile-source emissions from evolving light-duty vehicle fleets. However, ongoing studies will need to be maintained to keep such projections valid as new vehicle technology is introduced and older vehicles age.

While the great bulk of remote-sensing data available are for light-duty vehicles, emissions from heavy-duty diesel vehicles have been characterized, even though many North American heavy-duty diesel vehicles have substantial exhaust emissions that are not well sensed by normal low-level cross-road optical technology. Both traditional non-dispersive systems (Bishop et al., 2001a) and TILDAS systems (Jiménez et al., 2000b) have been deployed to reliably characterize onroad heavy-duty diesel trucks. These vehicles have emission distributions that are much closer to normal than the $\gamma$-distributions found for light-duty vehicle emission distributions. The same technology also has been used to characterize significant offroad mobile sources, such as snowmobiles (Bishop et al., 2001b). Measurements on a wide variety of offroad vehicles are underway and will help inform offroad mobile emission models.

It is possible to construct light-duty vehicle emission inventories directly from cross-road remote sensing data. Singer and Harley (2000) have used remote-sensing data to construct a fuel-based inventory for mobile emissions in Los Angeles, California, and Stedman and co-workers have recently published one such inventory for the Denver, Colorado metropolitan area (Pokharel et al., 2002). If more continuous and comprehensive remote-sensing data sets become available, the derivation of mobile emission inventories directly from these data may become widespread. However, because remote-sensing measurements typically sample each vehicle for less than a second, thus sampling a very small segment of each vehicle’s operating range, Wenzel et al. (2000) caution that it is necessary to sample a very large number of vehicles to estimate valid mobile-source emission inventories.

Remote-sensing measurement data have demonstrated utility for evaluating the effectiveness of air quality control programs. One of the earliest uses of remote-sensing data was to assess the impact of using oxygenated fuels to reduce mobile CO emissions (Bishop and Stedman, 1990). Systematic analyses of multi-year data sets have been used also to evaluate the effectiveness of I/M programs (Stedman et al., 1997). A recent extensive roadside pullover study that stopped vehicles which remote-sensing instruments had identified as high emitters and subjected them to conventional tailpipe emission inspections has confirmed that remote sensing does generally correctly recognize vehicles with excessive emissions (BAR, 2001). A recent report by the NRC concluded that remote-sensing measurements are an excellent source of onroad CO and HC emission data, that they also can be a useful screening tool to identify vehicles likely to pass or fail conventional I/M program tests, and that remote sensing is underutilized in current I/M programs (U.S. EPA, 1998; NRC, 2001). Since the impacts of control strategies must be factored into mobile-source emission inventories, remote-sensing data can play a key role in updating current emission inventories and projecting future emissions.
From a mobile-source standpoint, remote-sensing technology is now sufficiently developed that routine operational deployment as part of state or provincial I/M programs is feasible. The potential adoption of operational “clean screen” and/or “gross emitter” onroad I/M programs may provide nearly continuous and real-time remote-sensing data sets, which can be incorporated to keep mobile emission inventories much more current and provide better spatial resolution. Several large studies of commercial cross-road remote-sensing technology of ongoing I/M and other clean-air initiatives have been concluded recently (McClintock, 2002; ESP, 2003a; McClintock, 2004), opening up the prospect of more widely distributed and more continuous data sets gathered on a daily basis. For instance, a recent program sponsored by Missouri to test a clean-screen component for the I/M program for the St. Louis metropolitan area operated 20 to 26 days per month, collecting 300,000 to 500,000 vehicle-emission records per month for a total of nearly 5 million over the course of a year (McClintock, 2002).

Traditionally focused on VOCs, NO\textsubscript{x}, and CO, future mobile-source sensing technology can be expected to address additional chemical species during future years. There is an increasing concern about mobile sources of airborne toxic air pollutants, with particular emphasis on possible carcinogens including formaldehyde, acetaldehyde, benzene, acrolein, and 1,3 butadiene, as well as the organic portion of exhaust fine PM. Advanced remote sensing systems employing dispersive spectrometers (Baum et al., 2000) or tunable lasers (Nelson et al., 1998) have the capability to quantify some exhaust toxic air pollutants directly and may well be able to quantify related indicator compounds for others. Advanced remote sensing systems also have the capability to quantify mobile emissions of greenhouse gases beyond CO\textsubscript{2}, including N\textsubscript{2}O and CH\textsubscript{4} (Baum et al., 2000; Jiménez et al., 2000a). Both types of advanced systems have demonstrated the ability to quantify the production of NH\textsubscript{3}, an important PM precursor, on overactive NO reduction catalysts (Baum et al., 2000; McManus et al., 2002).

Remote sensing data for mobile sources are primarily reported on a fuel-use basis (g emissions/g fuel used). To be compatible with the current vehicle emission models, the fuel-based estimates are converted to g/VMT using estimated relationships between driving patterns and fuel consumption. Fuel-based emission estimates are preferred by some workers for evaluation of mass emissions and for speciation profiles for emitted PM. Fuel-based methods also offer important opportunities to evaluate transportation sources on a comparable basis with nonroad sources, which may become increasingly important for some applications.

### 6.1.2 Alternate Methods for Mobile-Source Characterization

Mobile-source emissions continue to represent one of the more challenging aspects of emission rate estimation. Typical mobile-source emission models such as the MOBILE series are idealized, in the sense that they assume standard driving cycles and fleets having uniform characteristics based on certification and dynamometer testing (e.g., Sawyer et al., 2000), leading to serious concerns regarding their “real-world” applicability. The models have received ad hoc adjustments for urban/rural situations as well as for Mexican and Canadian driving conditions, but remain relatively coarsely evaluated. In recent years, testing under real-world driving conditions has increased, using different creative approaches, which attempt to avoid limitations embedded in the existing emission models. One such approach –remote sensing – has been discussed in Section 6.1.1. Several additional approaches are summarized in the following subsections.

#### 6.1.2.1 Roadway Tunnel Studies

Roadway tunnel studies represent an additional method for testing motor-vehicle emission models, at least over limited ranges of conditions. These studies include gas and particle sampling from tunnel entrance and exhaust air using conventional instruments, documentation of driving conditions during sampling, documentation of the types of vehicles passing through the tunnel, and estimation of emission rate distributions expected from the fleet observed in terms of speed (or speed variation) and tunnel length. The first experiment of this type was conducted in the 1970s (e.g., Pierson and Brachaczek, 1983). More recently, several of these studies have been reported in Los Angeles, Baltimore
and Vancouver (e.g., Pierson, et al., 1996; Gertler et al., 1997; Pierson et al., 1996; Sawyer et al., 2000; Graham et al., 2003).

Tunnel studies have been useful in providing data for checking the reliability of models such as MOBILE and EMFAC to calculate traffic aggregate emissions of CO, VOC, NO\(_x\), and PM under a limited range of onroad conditions. Historically, these studies raised serious questions regarding the performance of emission models as early as the 1980s.

While the tunnel studies have limitations in themselves, they nevertheless are valuable for identifying ambiguities in model calculations, and have resulted in improved model estimates of gaseous emissions. Tunnel studies have been criticized for not being representative of a wide range of expected onroad driving conditions (including cold starts, and transients—fuel rich operation), for having ambiguities in vehicle operating conditions, and for having only limited representation of traffic mixes. They also give ambiguous results for evaporative emissions. Critics also have questioned whether or not the observed pollutant mixtures are characteristic of open-air conditions, given the constrained air circulation present in tunnels.

Despite these limitations, tunnel studies have been instrumental in providing cross-checks for mobile emission models (e.g., Sawyer et al., 2000). Partly in response to tunnel-study results, considerable effort has been devoted to improving the MOBILE and EMFAC emission models since the 1980s.

6.1.2.2 Mobile Laboratories and Chase Vehicles

The development of robust, fast-response sensors for many gaseous pollutants, as well as PM physical and/or chemical properties, has allowed the deployment of useful instrument suites in a variety of onroad vehicles. This combination enables truly “mobile laboratories” capable of real-time measurements while in motion. In general, modern mobile laboratories can be used to characterize onroad, mobile-source emissions in two distinct modes—vehicle fleet and vehicle tracking, or “chase” measurement.

In the fleet mode, mobile laboratories characterize onroad pollutant emissions in two ways. The most accurate and informative method is by intercepting individual vehicle exhaust plumes and correlating target pollutant enhancements with above-background CO\(_2\) levels. Less quantitatively, elevated onroad pollutant concentrations can be measured and correlated with traffic volume or average elevated CO\(_2\), or CO as an emission marker without resolving individual vehicle plumes. These fleet methods can be thought of as tunnel studies without the tunnel. Like tunnel studies, they yield a sample of fleet-averaged emission indices for individual pollutants. The individual plume-intercept method has the advantage of also yielding a full distribution of emission indices for the target pollutants, since an individual emission index is obtained for each exhaust-plume intercept. Since plume excess CO\(_2\) can be readily related to the fuel consumption rate through the combustion equation, measurements of emission ratios (plume excess pollutant/plume excess CO\(_2\)) can be directly converted to emission indices (g pollutant emitted/kg fuel consumed for different driving conditions).

In the chase mode, mobile laboratories are designed to sample exhaust plumes from specific target vehicles repeatedly. This mode can be used to characterize the emissions of either cooperative or non-cooperating “wild” vehicles over a range of operating parameters. The chase laboratory must shadow the target vehicle and must contain fast response (~1-s) sensors, typically measuring CO\(_2\) and the target pollution of interest. Data from fast-response sensors for the target pollutants are correlated with the CO\(_2\) data to yield emission ratios and derive emission indices for target vehicles as a function of operating conditions. The target vehicle’s speed and acceleration can be obtained from onboard sensors (for cooperative vehicles), or from the speed and acceleration values of the chase vehicle, plus range-finder measurements providing the instantaneous distance between the chase and target vehicles. Chase-vehicle measurements are similar to onroad dynamometer measurements, but typically may sample a wider range of operating conditions.

Fleet emission ratios for N\(_2\)O emissions from U.S. vehicles have been reported by Jiménez et al. (2000) and formaldehyde emission ratios for vehicles in Mexico City and Boston by Kolb et al. (2004). Jiménez et al. (2000) demonstrated that
the distribution of U.S. fleet N₂O emission ratios measured using onroad mobile laboratory plume sampling was very similar to that obtained by cross-road remote sensing, using tunable infrared laser differential spectroscopy sensors in both cases. A variety of mobile laboratories has been developed recently and deployed to characterize onroad pollutant levels and mobile emissions in Europe and North America (Seakins et al., 2002; Bukowiecki et al., 2002; Kittelson et al., 2004; Weijers et al., 2004; Gouriou et al., 2004; Kolb et al., 2004), although only the latter group has published fleet-emission ratios and indices based on ensembles of individual exhaust-plume emission ratios (Jiménez et al., 2000; Kolb et al., 2004). A number of groups have focused on characterizing onroad exhaust emissions of PM, with some placing particular emphasis on ultrafine or “nanoparticle” concentrations and properties (e.g., Kittelson et al., 2004, Gouriou et al., 2004; Weijers et al., 2004).

Kittelson et al. (2000) have deployed a “mobile emission laboratory” mounted in a cargo container on a truck chase vehicle to characterize onroad diesel emissions from cooperative heavy-duty diesels. Vogt et al. (2003) have instrumented a chase van to characterize emissions from cooperative light-duty diesel vehicles on a test track. Kolb and co-workers have utilized a large van-based mobile laboratory to quantify a range of gaseous and PM emissions from a range of heavy-duty diesel and heavy- to light-duty gasoline vehicles in Mexico City and several U.S. cities (Shorter et al., 2001; Cangaratna et al., 2004; Kolb et al., 2004). Initial analyses of these measurements indicate that the data are comparable to cross-road remote-sensing data, giving the advantages that individual vehicles can be sampled for a larger range of operating conditions and a much wider range of gaseous exhaust species and fine particle properties can be specified. For instance, the mobile laboratory described in Kolb et al. (2004) is equipped to quantify exhaust emissions of gaseous CO, NO, NO₂, HONO, NH₃, H₂CO, CH₃CHO, CH₃OH, benzene, toluene, C₂-substituted benzenes, and SO₂, as well as a range of PM properties, including number density, size distribution, and mass loadings of SO₄²⁻, NO₃⁻, NH₄⁺, OC species, and PAHs.

6.1.2.3 Portable Emission Measurement Systems

As their name implies, portable emission measurement systems consist of analytical equipment that is sufficiently compact and field-operable to allow deployment on vehicles under actual operating conditions. This is particularly important for some mobile sources – such as construction and farm equipment – which are difficult and expensive to subject to standardized testing. Portable emission measurement systems provide a way of testing such equipment without having to either remove the machine from service or modify it in any way.

Portable emission measurement systems have been under development for about a decade. The U.S. EPA, for example, recently patented the Real-Time OnRoad Vehicle Exhaust Gas Modular Flowmeter and Emissions Reporting System, better known as ROVER (U.S. EPA, 2003). As the first generation of portable emission measurement system devices, ROVER established a method for measuring mass flow from engines.

U.S. EPA workers have continued portable emission measurement system development and introduced the Simple Portable Onboard Test (SPOT) in 2001. SPOT was designed specifically for nonroad applications. Building on the ROVER system, the SPOT system consists of a rugged, compact package and provides further advances in mass-flow measurement and activity data capture. The technology has been licensed to equipment manufacturers and a competitive market has emerged to design and manufacture portable emission measurement systems that meet stakeholder needs.

Portable emission measurement system-related goals are to encourage and support private industry in equipment development and refinement. The U.S. EPA is also working on developing and demonstrating PM mass measurement. Prototype equipment is under evaluation in U.S. EPA and contractor laboratories to establish correlation between reference methods and prototypes. Development and field adoption of this capability is ongoing, and similar work will pursued for measuring toxic air pollutants in the future.
Currently, portable emission measurement systems are being deployed in a number of ways, such as checking compliance of in-use heavy-duty vehicles. The U.S. EPA is also undertaking an extensive program in Kansas City in 2004/2005 (Baldauf et al., 2004) to evaluate PM emissions from light-duty vehicles. About 500 cars are being tested on a portable dynamometer and most of those also will be equipped with portable emission measurement systems and returned to their owner for real world emission measurement. This will constitute the first large database for real-world, in-use emission measurements ever assembled. In addition, the U.S. EPA plans to launch a test program involving 150 nonroad engines. Plans are also in progress to test heavy-duty trucks.

Portable emission measurement systems enable the testing of motor vehicles in any location, and the cost of testing per vehicle is believed to be a fraction of that for laboratory testing – by an order of magnitude or two in the case of nonroad engines and heavy-duty trucks. These two advantages will allow testing of statistically significant national samples of vehicles. Portable emission measurement systems are believed to be sufficiently inexpensive that state governments can afford to acquire and deploy them to generate local emission and activity data for use in refined local and mesoscale modeling. This will improve the quality of data input for emission models such as MOVES, and transportation inventory projections.

6.1.2.4 Onboard Sensors

A complement to other mobile source measurements has been developed using the monitoring of onboard engine diagnostic (OBD) sensors in contemporary vehicles, such as exhaust-gas oxygen and temperature sensors, and engine-load and fuel-consumption monitors. By using an engine performance model in conjunction with a wireless device to communicate, these sensors can provide an essentially real-time, indirect estimate of CO, NO\textsubscript{x} and VOC emissions from equipped vehicles. Tests in California on a fleet of 1000 taxicabs, for example, have provided useful data on a large number of vehicles for comparison with the federal OBD II I/M emission test (e.g., Banet, 2003). Using these data, nonperforming vehicles can be identified, and returned for maintenance to correct failing emission control equipment. The data have not been used extensively as yet for comparison with other onroad measurements. Investigation of their application to emission estimation should yield at least qualitative performance data to check limited sampling data acquired from portable emission measurement systems and roadside monitoring.

In the future, new micro-sensors are under development that can withstand the conditions present in high-temperature, hostile post-combustion environments (e.g., http://www.es.anl.gov/html/sensor.html). The Argonne National Laboratory, for example, is developing prototype “smart” voltammetric/electrocatalytic microsensors combining cermet materials, voltammetry, and neural network signal processing. Tests have been conducted on these prototypes for CO\textsubscript{2} detection, but the sensors also can be “trained” to detect other chemicals, including VOCs. With these sensors, one can foresee opportunities for onboard emission sensing by vehicles far in advance of current practices. Such sensors also should have important applications as CEMs for a variety of industrial applications.

6.1.2.5 Sampling and Dilution Tunnels for Reactive Emissions

Although applicable for evaluating both stationary and mobile sources, sampling and dilution tunnels are of particular interest in the context of transient emissions from diesel-powered roadway vehicles, and thus are discussed here. Determining emission rates from sources whose pollutants transform quickly once emitted to the atmosphere has received increasing attention during recent years, primarily as a consequence of heightened importance placed on ultrafine PM and associated health impacts, as well as the evolution of specific combustion technology categories (e.g., advanced diesel engines), which emit large numbers of particles in this size range. Characterizing PM emissions from combustion systems is difficult because of the high temperatures and moisture content of exhaust gases, as well as the strong coagulative tendency of ultrafine PM in high concentrations. Upon exiting the stack the combustion products cool rapidly and dilute with ambient air, during which time physicochemical reaction processes such as condensation, nucleation, and coagulation change PM size distribution and composition. Measurement of PM in hot exhaust is
further complicated by the presence of semivolatile material, which may either undergo homogeneous nucleation or condense on existing particles, thus changing chemical composition and size distribution. Because particle count is dominated by the small particles, particle-number distributions are especially sensitive to sampling and dilution conditions. Measured particle-number concentrations can be changed many orders of magnitude by varying sampling conditions.

An obvious approach to this issue is to utilize fast-response instruments to continuously measure flow rates and the concentrations of species of interest. Frequently, this is not possible because of sample temperature, humidity, and the lack of suitable instrumentation. Dilution sampling (Lipsky et al., 2002) represents an alternative technique for this purpose. Depending on their design, dilution samplers either quench the physicochemical reactions by rapid dilution and cooling, or else they provide for quantitative examination of these processes by mixing with specified amounts of conditioned air.

Some sampling systems have added residence chambers to increase the time between exhaust dilution and sampling the diluted emission stream (e.g., England et al., 2005). This is done to allow additional time for equilibration between the semivolatile constituents and the PM as well as time for coagulation of some of the fine PM. Additional dilution of the sample immediately before the residence chamber can be done to better simulate the effluent after equilibration with ambient conditions. While dilution tunnels are designed to minimize particle losses, sampler surface interactions between the gases and particles remain problematic. Care has to be taken in the sampling system to determine losses of trace reactive gases and particles. Semivolatile materials tend to slowly adsorb and desorb from tunnel walls, resulting in a variable background contribution from the tunnel itself. Hence, the tunnel must be conditioned carefully if it is used on sources of greatly varying magnitude.

One of the problems associated with the use of dilution tunnels is their lack of portability for source sampling. Many sources are large and involve equipment or stacks that are at substantial heights. Dilution systems designed in the 1980s and later generally are bulky and clumsy to transport. Recently, designs based on these large units have been built, with residence times needed for equilibration much shorter than the larger units. England et al. (2005), for example, have designed a new portable tunnel based on much larger, bulkier units of Hildemann et al. (1989) at the U.S. EPA and the Desert Research Institute. This and other portable dilution tunnels have been demonstrated for sampling of stationary sources. The England et al. design has been used for mass emissions and speciation analysis of PM$_{2.5}$ from a selection of gas- and oil-fired combustors. The results also provide important data and insight into emission behavior, as well as ultrafine PM and semivolatile material from these sources. Based on the performance testing of dilution-tunnel units for stationary sources, an ASTM committee has been formed to codify the sampler and sampling method (ASTM D22.03/W1752 certification). This method will probably replace U.S. EPA standard methods 201A and 202 in 2005.

Tests for PM emissions from stationary sources using dilution samplers have shown significant differences compared with Methods 201A/202 (e.g., England et al., 2002). In testing of effluents from a natural-gas process heater and boiler, for example, the proposed ASTM method indicates that the dilution tunnel samples yield much lower PM concentrations from such combustors compared with Methods 201A/202, and from estimates based on default values in the AP-42 guidelines. Thus, the sampling method for source effluents of PM clearly depends strongly on the suitability of sampler design. This will significantly influence certain published emission factor estimates currently used in practice.

Fast-response sensors, fast-response mass flow controllers, and improved electronics are likely to result in more accurate and reliable dilution samplers in the near term. The impact of semivolatile material on PM measurement, however, will not be solved by improved sampling methods. Standardization of measurement procedures should greatly increase precision of the measurement, but will not resolve the problem of applicability to different atmospheric conditions (i.e., ambient temperature, humidity, dilution, and background PM). Relating sources of rapidly reactive emissions to ambient concentrations will require (a) improved modeling of atmospheric
processes, (b) innovative statistical inferences to relate measurements of a few samples to conventional averaged estimates, and (c) more geographically and seasonally comprehensive source measurements of the appropriate parameters as input to models.

6.1.3 Continuous Emission Monitoring Systems

CEMS have been described in some detail in Section 4.7.4, and are mentioned again here mainly to note that these systems, and their application, can be expected to evolve substantially as newer sensors are developed (e.g., Jahnke, 2000). As discussed in Chapter 4, CEMS for SO$_2$, NO$_x$, CO$_2$, O$_2$, opacity, and flow have been deployed on large electric utility boilers in the United States since the early 1990s to comply with the allowance trading requirements established by the Acid Rain Program under Title IV of the U.S. 1990 Clean Air Act Amendments. Hourly emission data from CEMS are reported quarterly to the Acid Rain Program’s Electronic Data Reporting System. This information forms the basis for the annual emission data included in the U.S. emission inventories for electrical generating units and also serves as highly accurate input to modeling inventories. The variations in emissions recorded by CEMS reflect changing boiler and combustion-turbine operating conditions, fuel compositions, meteorological conditions, start-ups, and shut-downs. Additional continuous NO$_x$ monitors will be installed on other sources in the eastern United States in the next several years to comply with allowance trading that is integral to the NO$_x$ budget program. In addition, opacity monitors are commonly installed on incinerator stacks.

CEMS for gas-phase constituents typically use optical sensors, based on absorption, emission, or fluorescence, depending upon the species being detected. Pollutant emission rates are calculated by multiplying pollutant concentrations by stack volumetric flow. Table 4.5 contains a representative list of mature and developing CEMS technologies indicating the physical measurement basis for each pollutant species.

Many challenges are associated with demonstrating the accuracy, precision, and reliability of CEMS. Substantial effort will be needed to develop protocols and experience to ensure that their operation provides credible data. For example, interferences, such as those associated with UV absorbance bands of SO$_2$ and mercury, must be minimized. Precision and accuracy at low emission levels, such as single-digit ppm NO$_x$ levels from state-of-the-art combustion turbines must be demonstrated. CEMS must be maintained and carefully calibrated in hostile thermal and corrosive atmospheres of hot effluent gases. Validation and management of a large body of continuous data from thousands of sources presents a formidable task for the operators and for the U.S. EPA and states as the archivers of these data. In spite of these difficulties, Section 7 demonstrates that CEMS systems on power plants have yielded highly accurate emission measurements for NO$_x$, SO$_2$ and CO$_2$.

CEMS for other pollutants are being developed in the United States and Europe for regulatory compliance, for process-control needs, and to support future allowance-trading programs. Pollutants for which CEMS are currently under development include NH$_3$, BC, mercury (total and speciated), PM, and VOCs. Most of the development is being conducted by instrument manufacturers with the expectation of growing markets for this technology.

6.1.4 Aircraft Plume Measurements

An important alternative to remote sensing of stationary sources is the use of aircraft, helicopters and even blimps to sample plumes, and characterize near-stack chemical reactions with distance downwind. Plume tracing has been used to characterize, at least qualitatively, gas and PM emissions from large power plants, industrial sources and urban areas since the 1970s (e.g., Easter et al., 1980; Cher et al., 1984).

In the past, emission rate determination using aircraft sampling has been difficult because of large uncertainties in identifying the plume location and the fluxes of pollutants moving through plume cross sections. One of these problems derives from the lack of fast-response instruments compatible with aircraft speeds. Another arises from the imprecise sampling and measurement from an aircraft flying at different altitudes. Yet another is the lack of complete, simultaneous measurements that provide...
for estimating a material balance for reactants and products. Recently, plume measurements have become more quantitative with the use of fast-response instrumentation, global positioning system (GPS)-based position measurement, and carefully planned, systematic sampling across plumes (cf. Chapter 7).

Section 7.3.2 discusses an extensive set of plume flux determinations that are used to evaluate inventoried power plant emissions. In general, quite good agreement is found between the aircraft flux measurements and the fluxes derived from CEMS data. Attempts to quantify emissions from urban areas are described by Trainer et al. (1995), Klemm and Ziomas (1998), and Plummer et al. (2001).

A major challenge in these measurements is developing and deploying instruments that can perform fast response (~1-s resolution) measurements of the ambient concentrations of the emitted species with sufficient accuracy and precision. In addition, the wind speed and direction at the time of emission are required to derive a flux from the concentration measurements. Current instrumentation is adequate for fast response measurements of CO, NOx, SO2 and CO2. Biomass burning emissions have been determined by Fourier transform infrared spectrometry and other techniques that often have considerably slower response times. In this case various integrating procedures are utilized (Sinha et al., 2003). However, much of this current instrumentation is heavy and requires a good deal of electrical power and experienced operators. It could be more extensively applied if lighter, lower powered and more routinely operated instruments are developed. There is an important need to develop instrumentation for determination of speciated VOC emissions from intense sources such as petrochemical industrial facilities, as discussed in Section 7.3. Such emissions have been identified as critically important to some outstanding air quality issues, for example, in developing Texas state implementation planning for achieving the ozone standard.

### 6.1.5 Direct Flux Measurements

In the context of the present section, “flux measurements” pertain to direct or indirect determinations of pollution fluxes (amount of pollution issuing from a unit area of the Earth’s surface per unit time) occurring by vertical turbulent transport within the Earth’s boundary layer. Measurements of this type typically apply to emissions from sources having relatively uniform spatial distributions (e.g., isoprene emissions from a forest canopy, NH3 emissions from agricultural land, dimethyl sulfide emissions from an ocean surface) or aggregated sources which can be approximated as uniform in a spatially-averaged context (e.g., urban and suburban areas under specialized conditions). Because pollution fluxes can be either positive (emission) or negative (deposition), measurement technology for emission flux measurements closely parallels that for dry-deposition assessment.

Although several inferential techniques have been applied to estimate pollution fluxes (Hicks et al., 1987), most direct emission flux measurements fall into two basic classes: profile measurements and covariance methods. As their name implies, profile measurements depend on observations of vertical pollutant profiles in conjunction with appropriate meteorological measurements. Typically fluxes are calculated on the basis of these observations using an equation of the form

\[
\text{flux} = -K_z \frac{dc}{dz}
\]  

where \(\frac{dc}{dz}\) is the vertical gradient of pollutant concentration, and \(K_z\) is a vertical transport coefficient inferred from meteorological observations.

Covariance methods (Rinne et al., 2001) measure fluctuations in local concentrations and wind-velocities, and calculate fluxes on the basis of fundamental turbulence theory using the form

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4 Enclosure methods, wherein an emitting area is enclosed in a monitored chamber, constitute another class of flux measurements (cf. Baldocchi et al., 1996). This class is of more limited applicability in the context of criteria pollutants and their precursors, and will not be considered further here.
where \( w' \) and \( c' \) respectively are the fluctuating components of the vertical wind-velocity and concentration, and \( t_2 - t_1 \) is some appropriate time-smoothing interval. Measurement approaches that observe \( w' \) and \( c' \) directly and compute fluxes by subsequent integration often are referred to as eddy-correlation techniques.

Eddy correlation instrumentation must be sufficiently responsive to monitor all important components of the turbulence spectrum, typically frequencies of the order of 0.1 Hz and higher; and while modern wind-velocity instrumentation can satisfy this requirement easily, current pollution monitoring equipment often cannot. Eddy-accumulation methods, wherein the \( w' \) sensing device operates a fast-response valving system feeding two air sampling reservoirs at rates directly proportional to \( w' \), represent one method of overcoming this difficulty. Subsequent concentration measurements of the two chambers leads to evaluation of the integral in Equation (6.2). Additional attempts to overcome the time-response issue have led to various “relaxed” or “disjunct” covariance approaches, wherein the required sampling frequency is degraded in one form or another (Wesely, 1988). Such approaches typically involve additional approximations or limiting assumptions regarding turbulence behavior.

In addition to the obvious challenges posed to measurement instrumentation, both profile and covariance techniques depend strongly on measurement location, meteorology, and pollutant behavior. Equations (6.1) and (6.2) both are single, one-dimensional components of the general relationship describing three-dimensional, transient behavior in the real atmosphere, and their valid application for flux measurement generally requires that the local environment approximate a one-dimensional steady state. This in turn implies a uniform source distribution and a uniform wind fetch. Informed meteorological guidance is essential for determining the presence of these conditions. Moreover, while Equation (6.2) can be considered a fundamental, first-principles relationship, Equation (6.1) is more inferential in the sense that the transport coefficient, \( K_z \), is quasi-empirical in nature and is an estimated entity. This renders profile measurements somewhat more subject to uncertainty - in principle, at least - than their covariance counterparts.

Finally, Equations (6.1) and (6.2) both imply that turbulent transport dominates the total flux term, a situation that will be violated if other mechanisms, such as gravitational sedimentation, are significant. In addition, chemical reaction of the observed component, if sufficiently rapid, can confound both profile and covariance observations.

Direct flux-measurement techniques have been applied for both monitoring and special-purpose measurements. Table 6.2 provides several examples of such observations. As noted above, both profile and covariance methods are limited by applicable source and meteorological conditions. Covariance methods tend to be less demanding in this respect, and this combined with their stronger theoretical basis suggests their preferential use during future years.

The currently rapid development of sensitive, fast-response chemical instrumentation (e.g., mass spectroscopy) will encourage extensive future covariance-method application, both on stationary and aircraft platforms. While the bulk of these applications will focus on relatively ideal boundary-layer situations, some encouraging developments are taking place in less ideal settings, such as urban and mixed suburban areas (Velasco et al. 2005; Fowler et al., 1997). Future results will determine practical applicability and the extent of future use under such situations.

The estimation of fluxes from diffuse ground level sources, for example windblown dust, has proven to be a major challenge for inventory development. Simulations by air quality models, which approximate vertical dispersion in atmospheric surface layers can overestimate dust emissions by a factor of two or more, compared with actual ambient measurements (e.g., NARSTO, 2004). To resolve this issue, substantial effort has been invested recently in developing improved methods for estimating such surface emissions, as reviewed by Watson and Chow (2002). Countess et al. (2002) also have reported...
A methodology for estimating windblown dust and resuspended road dust fluxes for application to regional-scale air quality models.

### 6.1.6 Summary of Measurement Alternatives

Table 6.3 provides a summary of the measurement methods discussed in this chapter and their application, as well as a conceptual summary of their relevance to emissions and activity factors. This listing is intended as a reference point for more detailed examination of the above discussion, as well as contributions to the published literature.

### 6.2 MODELING AND INTERPRETIVE METHODS

Over the last 25 years, air quality models establishing the relationship between sources and ambient concentrations or receptor exposure have improved dramatically for both nonreactive and reactive pollutants. In principle, the models can be used to estimate ambient conditions for periods of less than a day to multiple years, provided appropriate emission and meteorological data are available. Sufficient testing and evaluation of models has been accomplished over the years that workers have identified emission estimation as one of the major uncertainties in the model results (e.g., NARSTO, 2000; 2004).

Chemical-transport models have offered an important opportunity for checking the reliability of the emission models on which they depend. The methods use two different kinds of models that are readily available: (a) receptor-oriented modeling, and (b) source-based modeling. Recent advances increase the potential for both techniques. The following sections describe recent improvements in the reliability of emission estimation though insights provided by air quality model results.

#### 6.2.1 Receptor Modeling

Receptor modeling is a term describing a variety of (largely) statistically associated techniques for inferring source categories and/or magnitudes on the basis of ambient-concentration measurements. Differences among these various techniques are based on the types of statistical approaches employed and the types of observational patterns (e.g., temporal differences, spatial differences, ratios among specific compounds) considered, as well as whether the specific technique depends on source-profile information in addition to ambient measurements. Although most receptor models are totally statistical in nature, some variants employ limited deterministic calculations as well. Receptor modeling is most applicable to situations where differential attrition of the observed species, by deposition or by reaction, is minimal, although techniques to deal with such issues have been incorporated into some receptor models by adding quasi-deterministic components.

<table>
<thead>
<tr>
<th>Pollutants</th>
<th>Technique</th>
<th>Source/Location</th>
<th>Platform</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isoprene, CO₂</td>
<td>Disjunct eddy covariance</td>
<td>Deciduous forest Michigan</td>
<td>Tower</td>
<td>Westburg et al. (2001)</td>
</tr>
<tr>
<td>Ethane</td>
<td>Disjunct eddy covariance</td>
<td>Urban area, Mexico City</td>
<td>Rooftop tower</td>
<td>Velasco et al. (2005)</td>
</tr>
<tr>
<td>CO₂</td>
<td>Eddy covariance</td>
<td>Various locations throughout the world.</td>
<td>Tower</td>
<td>Baldocchi (2003)</td>
</tr>
<tr>
<td>Suspended dust flux</td>
<td>Eddy covariance</td>
<td>Prototype testing in western United States</td>
<td>Tower</td>
<td>Gillies et al. (2003)</td>
</tr>
<tr>
<td>Method</td>
<td>Application</td>
<td>Example Metric</td>
<td>Emission Factor</td>
<td>Activity Factor</td>
</tr>
<tr>
<td>-------------------------------</td>
<td>------------------------------------</td>
<td>----------------------------------------</td>
<td>---------------------------------------------------------------------------------</td>
<td>---------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>Remote sensing (Ground based-ambient)</td>
<td>Stationary and mobile sources --cross plume observations.</td>
<td>Gaseous spectral absorption or PM light extinction.</td>
<td>EF estimate from process data and emissions as a function of time; short time comparison with direct stack tests.</td>
<td>Direct measurement of emissions as a function of time; with EF convert to activity estimate.</td>
</tr>
<tr>
<td>Remote sensing (Aircraft and satellite)</td>
<td>Stationary sources; intermittent-wildfires, dust storms.</td>
<td>Gaseous spectral absorption; PM light extinction.</td>
<td>Emission rate estimates compared with calculated long term EF; interpretation of large scale EF for natural sources.</td>
<td>Capability for estimating time and space variability of natural sources, and testing time variation of stationary sources.</td>
</tr>
<tr>
<td>Roadway tunnel studies</td>
<td>Mobile sources --controlled measurements constrained by the tunnel conditions.</td>
<td>Conventional gas and PM instruments.</td>
<td>Short term, traffic-based emission rates translated into EF.</td>
<td>Limited activity data based on tunnel constraints.</td>
</tr>
<tr>
<td>Mobile laboratory and chase vehicles</td>
<td>Mobile sources</td>
<td>Conventional gas and PM instruments; vehicle identification records.</td>
<td>Short term emission rate measures translated into EF from real world fleet.</td>
<td>Activity factors interpreted indirectly from traffic patterns and emission rates for vehicles.</td>
</tr>
<tr>
<td>Mobile onboard sensors</td>
<td>Mobile sources</td>
<td>New technology relying on link of gas microsensors and vehicle computer system.</td>
<td>Estimate of fuel based EF from detectors, combined with vehicle engine performance.</td>
<td>Time variation in emission rate enables estimate of activity factors given emission rate.</td>
</tr>
</tbody>
</table>
to account for these effects. Receptor modeling should be applied with special caution when such conditions are suspected, especially for regional and larger spatial-scale conditions.

As with inverse modeling, receptor modeling is generally considered more as a verification/reconciliation tool, as well as a means for generating semi-quantitative insights regarding questionable or unknown emission sources. The success of receptor modeling depends heavily on the selection and quality of associated input measurements. Moreover, receptor-model results can be strongly prone to misinterpretation unless treated with appropriate caution: The experience and capability of the practitioner is of considerable importance in this regard.

Receptor-modeling techniques are described in detail in several reviews (NARSTO, 2004; Chow and Watson, 2002; Henry et al., 1984; Henry et al., 1997; Hopke and Dattner, 1982; Hopke, 1999; Moro et al.,

<table>
<thead>
<tr>
<th>Method</th>
<th>Application</th>
<th>Example Metric</th>
<th>Emission Factor</th>
<th>Activity Factor</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dilution tunnel sampling</td>
<td>Stationary and mobile sources (New ASTM reference certification for stationary sources)</td>
<td>Sampling of emissions approximating real world conditions for dilution (gases and PM).</td>
<td>Emission rate determined with speciation for condensed phase, volatiles and semivolatiles; comparison with EF from conventional methods.</td>
<td>Time variation over short term sampling provides estimate of activity factor with emission rate variation.</td>
<td>Limited to short term testing of stationary sources, and dynamometer testing of vehicles; translation into long terms averages for emission estimation not well established - new reference method in process.</td>
</tr>
<tr>
<td>Continuous emission monitoring (CEMs)</td>
<td>Stationary sources</td>
<td>In-stack observations of a few key gases; opacity for PM semi-quantitative</td>
<td>Emission rate directly measured with stack flow rate and concentration; infer EF from data.</td>
<td>Activity estimated from time variation in emission rate; gives long term and short term variability.</td>
<td>Major improvement in emission rate of stationary sources based on gas measurements. Limited to large stationary sources.</td>
</tr>
<tr>
<td>Airborne plume observations</td>
<td>Stationary sources; intermittent sources like wildfires</td>
<td>Conventional gas and PM observations, with ancillary aircraft speed, location</td>
<td>Emission rate inferred from cross plume tracking; EF inferred if effluent rate known.</td>
<td>Activity factor potentially inferred from emission rate.</td>
<td>Limited in space and time; depends strongly on ability to calculate flux from plume cross-section.</td>
</tr>
<tr>
<td>Ground based-direct flux observations</td>
<td>Ground level fugitive sources</td>
<td>Conventional multiple gas and PM instrumentation using vertical stratification.</td>
<td>Emission rate estimated from vertical gradient observation or analogous means. Translated into EF using ancillary meteorological and land surface data.</td>
<td>Activity factor estimate based on time variation in emission rate, and ancillary observations.</td>
<td>Limited by undetermined representativeness of flux conditions for dust rise, or from vegetation; reliability of application to broad range of conditions unknown.</td>
</tr>
</tbody>
</table>
1997; Pace, 1991; Seigneur et al., 1999; U.S. EPA, 1984; U.S. EPA, 1985; Watson, 1984; Watson et al., 2002; Watson and Chow, 2004). Brook, Vega, and Watson (see NARSTO, 2004) provide an informative summary table, which itemizes the various receptor techniques and compares their input requirements as well as their strengths and weaknesses.

From a historical perspective, it is noteworthy that receptor modeling activities were the first to identify the need to extend emission inventories far beyond urban domains to estimate long-range sulfur transport (Hidy, 1994), that fugitive dust was identified as an important emitter (Gatz et al., 1981), that residential wood burning emissions needed to be inventoried (Watson, 1979), that meat cooking emissions needed to be inventoried, that cold-start emissions were not accounted for in inventories (Watson et al., 1998), and that CO and hydrocarbon emissions were underestimated by emission models (Fujita et al., 1992, 1994, 1995). Many of these sources were previously omitted from local emission inventories, so source-oriented modeling existing at the time was incapable of accounting for their contributions.

Emission identification and reconciliation using receptor-model applications for atmospheric pollution source analysis are presently at a relatively mature state. Although such models can expect to experience further development during future years, they can be considered to be a stable cadre of useful tools for emission verification and analysis, which should see continuing and extensive future application.

**6.2.2 Inverse Modeling**

Inverse modeling involves data-analysis procedures that employ a deterministic source-receptor model to derive information on sources (which are usually considered as independent model variables), based on sampled values of observable, dependent variables such as ambient pollutant concentrations. In a somewhat simplistic sense, conventional atmospheric source-receptor models adhere to the general form

\[ c_i(x, y, z, t) = f(Q_1, Q_2, Q_3, \ldots, v_1, v_2, v_3, \ldots) \]  

(6.3)

where the dependent variable \( c_i \) represents concentration of pollutant \( i \) at some point \( x, y, z \) and time \( t \), the \( Q \)'s denote strengths of associated emission sources, and the \( v \)'s the remaining ensemble of independent model variables such as winds, solar insolation, and deposition rates. Inverse modeling for source evaluation involves a reformulation of the modeling equations to express the \( Q \) variables (usually implicitly) in terms of the observed concentrations, combined with the remaining independent variables. This essentially involves “running the model backwards,” hence the term “inverse modeling.”

The mathematical details associated with inverse modeling are described in a number of texts (e.g., Bennett, 2002). To date inverse-model applications have been much more extensive in non-atmospheric areas such as ground-water contaminant transport and seismic analysis (e.g., Parker, 1994; Zheng and Bennett, 2002), although significant recent advances have occurred in the atmospheric field (Bennett, 2002; Enting, 2002).

Although inverse modeling is currently considered a potentially useful top-down verification of existing, bottom-up emission inventories, it is not generally viewed as a direct tool for quantifying specific emission sources. There are two major reasons for this. First, the source-receptor models applied for inverse analysis are subject to substantial overall (i.e., global) uncertainty, owing to stochastic considerations as well as to uncertainties in model characterizations of individual contributing phenomena. Emissions calculated from inverse models on the basis of sampled concentration values are typically highly sensitive to this global uncertainty. Second, the sampled concentration data applied for inverse-model analysis must be chosen judiciously, and one must be careful to collect a sufficient number of samples to provide a well-specified mathematical system. Information redundancies between data samples – which are usually difficult to evaluate in a direct manner – add to the complexity of this specification problem. It is apparent that future model improvements combined with accumulated application experience will render inverse-modeling applications more quantitative during future years.

Inverse-model applications for atmospheric source analysis have been confined mainly to large-scale phenomena and disperse sources, such as global
CO₂ emissions (Gurney et al., 2002), global CO emissions (Pétron et al., 2002; Palmer et al., 2003a,b; Kasibhatla et al., 2003), and continental NH₃ and carbonaceous PM emissions (Gilliland et al., 2003; Park et al., 2003).

As noted above, atmospheric inverse-modeling techniques have lagged those in the other geological sciences. Improvements in atmospheric models combined with more general appreciation of the technique’s potential for emission analysis, however, will undoubtedly result in significantly increased application during future years. It can be anticipated that future intensive field studies will be designed with direct inverse-model applications in mind, resulting in collection of data specifically intended for this purpose.

6.2.3 Specialized Field-Study Design

The use of models to evaluate emissions, and the application of specialized measurements, are optimized by a careful design for field studies. Experience in developing appropriate designs for verifying emissions from different sources has expanded dramatically in the last decade or so. Field studies have become increasingly expensive, yet there are significant opportunities to conduct meaningful experiments with relatively simple and inexpensive approaches. Perhaps the simplest situation concerns the emissions from a single source. Complexity increases substantially when multiple sources of individual species are present.

Ideally a source-verification design should include:

- At least qualitative specification of the chemical constituents emitted by the source, and identification of similar sources in the surroundings that may confound the emission estimation.
- Specification of the time duration for estimating the emission rate, determination of the expected material input and process variables (e.g., fuel composition), and notation of the availability of test data from this or a similar source.
- Identification of appropriate sampling and instrumentation, including response times, specificity of composition, etc. to determine or estimate the emissions for the period of interest.
- Consideration of ancillary tracer measurements to assist in tracking the plume a posteriori.
- Consideration of the chemical reactions that may take place between the exit point and downstream tens of meters.

Few source-oriented experiments aimed at establishing emissions have taken all these requirements into consideration. However, a number of recent studies have been designed to account for some of the elements listed above. Notable among the design of source-oriented experiments recently are hybrid-source receptor studies including the investigation of the Mojave power plant in southern Nevada (e.g., Eatough et al., 2000), the Hayden and Craig power plants in west-central Colorado (Watson et al., 1996), the Nashville urban and power plant studies (e.g., Ryerson et al., 1998), the BRAVO experiment (Pitchford et al., 2004), and TexAQS2000 (2003). These studies were not designed to obtain emission rates per se, but emission rates were a necessary element for the studies to examine dispersal of the plume and its impact on surrounding areas, including human exposure, and visibility degradation.

6.3 ADVANCED DATABASE MANAGEMENT

In Chapter 2, a vision for improved database management was described. This vision includes applying innovative methods from areas outside of pollution engineering technology. While applying new methods from outside the field will take place over time, there are a number of opportunities to apply conventional database management approaches today. This section summarizes these opportunities.

Emission inventories like the U.S. NEI contain very large data collections. The NEI alone, for example, contains more than 60 million items. Access to the inventories now involves complex manipulations of data, which remain user “unfriendly.” A major challenge for managers of emission data, along with producing quantitative uncertainty estimates,
is the development of a means for access that uses contemporary management techniques, accommodates a variety of different data sources, and ensures data quality. This challenge is compounded by the need for efficient access of large amounts of spatially and temporally resolved data for air quality modeling. Access to these collections depends not only on efficiency of entering the tabulations for specific locations and times, but also on the ability to summarize emissions in terms of sources and emission density graphically, or by other visual means.

Most current emission databases are not seamlessly integrated with the other information sources needed to design emission control strategies. For example, the task of de-trending air quality data to isolate the effect of meteorological fluctuations and year-to-year emission changes is made extremely cumbersome by the data architectures currently used by regulatory agencies. The problem is further compounded by the fact that the emission databases typically are not integrated with geographically encoded land-use and urban-planning information systems. There has been an explosion in the use of GIS by urban planning agencies; but these advances are only beginning to be incorporated into the systems used by air pollution agencies. Moreover, there is a distinct need for standardized electronic, possibly Internet-based, data-reporting systems, which will allow industries and state/local/tribal agencies to report data, make emission estimates, and perform quality-control checks online.

When designing the data architectures and reporting systems it must be recognized that inventories serve multiple purposes. One of the reasons why there are no real incentives to improve the quality of the existing data is that the local agencies charged with the permitting process do not derive any real benefits from the current systems that are used to aggregate emissions for air quality modeling studies. Once the local data are aggregated to the state and regional levels, there is no feedback to the permitting officers. Open access to local emission information would provide an incentive for both the public and industry to ensure that the information is correct. As previously discussed, collection and submittal of emission inventory information by agencies might be improved if federal or state funding is explicitly linked to these activities being completed.

In the future, it is expected that a North American emission inventory will be universally available to all who want to access its information. This facility should be capable of handling information that has high temporal and spatial resolution, is source and facility specific, is comprehensive with respect to pollutants and sources, is well documented, and is based on comparable methodologies and factors. North American emission inventories need to be cast in common formats and units, or transparent conversion between metric and English units should be built into the data management software. Advances in information technology and the pursuit of collaboration among emission inventory organizations are paving the way to an inventory that meets these goals.

6.3.1 Distributed Emission Inventory Network

The guiding principles of an integrated emission inventory follow those of distributed databases and distributed computing. The design objectives are to create a network of relevant data, as well as visualization and analysis tools, which is characterized by the following attributes:

* Distributed. Data are shared but remain distributed and maintained by their original inventory organizations. The data are dynamically accessed from multiple sources through the Internet rather than collecting all emission data in a single repository. Responsibility for data quality and availability remains with the data providers; however, data users also can make their collated and refined data sets available to other users via this distributed system.

* Non-intrusive. The technologies needed to bring inventory nodes together in a distributed network need not be intrusive in the sense of requiring substantial modifications by the emission inventory organizations in order to participate. However, there will need to be some harmonization of existing inventory data and
structures and assignment of certain functions to maintain a fluid system.

Transparent. From the emission inventory user’s perspective, the distributed data should appear to originate from a single database to the end user. One-stop shopping and one interface to multiple data sets are desired without required special software or download on the user’s computer.

Flexible/Extendable. An emission data network should be designed with the ability to easily incorporate new data and tools from new providers joining the network so that they can be integrated with existing data and tools.

Figure 6.4 depicts an envisioned end state of a distributed emission inventory. Distributed data sources (emission estimates, activity data, surrogates, etc) in a variety of formats (relational database-management systems, text files, etc.) are available through the Internet and registered in one or more data catalogs. These data can be uniformly accessed with the aid of data wrappers (translators) and connected with web tools and services to support a variety of end applications. Mediators are used to find and combine the appropriate mix of data and services to fulfill a user’s task.

Information technologies available to develop a distributed emission inventory include web services and related interoperability standards, such as the OpenGIS Consortium Web Map Server and Web Feature Server specifications. An important aspect of many of these technologies is that they can be implemented without requiring substantial modifications of the existing emission inventory databases and data dissemination systems.

6.3.2 Data Sharing and Web Services

Many emission databases are already accessible through Internet-based methods either through direct data-file download or through web query tools. The query systems allow users to filter and access data at multiple levels of detail. These systems were designed to meet the needs of individual end users who log in to the online system, manually complete forms for defining their query, and then view the results in tables/graphics or download the data. While these systems serve the individual user, they are not easily integrated into a distributed emission inventory framework where automated computer-to-computer, rather than human-to-computer access is needed. However, supplementing existing systems with distributed data capabilities is feasible.
Approaches to developing distributed emission inventory databases can be grouped into two general categories: those that make data files available for external access and those that implement innovative information technologies and standards to make their data dynamically accessible.

For smaller inventories the solution can be straightforward – provide an Internet (ftp or http) address where Microsoft Access, Microsoft Excel, or ASCII text files are stored, giving a description of the data format and a consistent file-naming convention. For example, if emission inventory A has point emission data for each year since 2000 and each year is stored in a separate file, they might use a naming convention such as NAME_Type_Year so that the files stores would be InventoryA_Point_2000.dat, InventoryA_Point_2001.dat, and so forth. An external user can then automatically access these data using a ‘get data’ request based on that inventory’s file naming convention. When new data become available in the inventory, the ‘get data’ request can automatically retrieve the new data.

For larger, more complex inventories, alternative methods are in order. Certainly, a larger inventory could provide subsets of its inventory in separate files through ftp or http addresses. A more attractive scenario is where the forms that allow users to query the inventory are “filled out” by the computer rather than by hand. This would allow dynamic access to the data and opens possibilities for “middleware” to provide value-added processes (filtering, aggregation, and integration) to the data. Middleware may make the data available in a specialized format, in different graphical views, or through tools that allow the data to be combined with other data. For example, a middleware application may provide a tool that could access multiple distributed inventories through a single query form thereby avoiding the need for a single user to access each inventory individually.

An emission network using middleware would allow automated access to each emission node on the network after each data server implemented a web interface method of dynamically accessing its data. A new category of information technology, called web services, offers one method for creating nodes on the distributed network. Web services consist of self-contained software that uses XML-based standards for describing themselves and communicating with other web resources. These characteristics allow web services to be reused in a variety of independent applications.

In the web services network approach, mediators serve the role of brokers, providing users with the interfaces for finding available data, dynamically retrieving it, and integrating it with other distributed data sources. These network users can function on an independent level, each addressing local issues of importance. These individual components can then be integrated or modified to handle differing data types dynamically on demand.

Web service technology is still evolving and does not currently provide a complete off-the-shelf software solution. However, many required components are considered standards in web programming applications and therefore make it possible to create an operational data web service network. These components allow computer-to-computer communication in a platform- and programming language independent manner. Additionally, web service technology provides existing software applications with service interfaces without changing the original applications, allowing them to fully operate in the user’s existing environment.

Distributed data network principles are being studied and implemented by a variety of emission relevant projects and programs:

- The CEC commissioned a study examining available data and technology for a distributed North American emission inventory (http://capita.wustl.edu/NamEN).
- The U.S. EPA is initiating a program for Networked Emission Inventories for Global Emission Inventories (http://www.neisgei.org).
A NASA- and NSF-funded project is pursuing web services for aerosol data and tools (http://www.datafed.net).

The U.S. EPA is developing a network for exchanging data between states and the agency (http://exchangenetwork.net).

A U.S. federal effort aims to provide single-point access to interoperable data sources (http://www.geo-one-stop.gov).

Other examples of Internet-based systems include CARB’s GIS mapping tool, recently developed to display the amounts and spatial distributions of emissions in California. The prototype system is called CHAPIS, Community Health Air Pollution Information System, and provides a mapping tool to analyze the spatial distribution of emissions from point, area, and mobile sources of various criteria and toxic air pollutants. In addition, the Great Lakes Commission has begun development of an Internet-based system to provide emission data, including charting and mapping tools designed to meet client needs. The system is called CAROL, Centralized Air emission Repository OnLine, and is designed to provide easy access to the Great Lakes Toxic Air Pollutant Emission Inventory.

The distributed emission inventory concept, combined with electronic formatting for data entry, and electronic access to AP-42 data is expected to enhance the timeliness of developing a bottom-up emission inventory like the NEI. In time, analogous methods are likely to be deployed in Canada and Mexico to assist the three nations in more rapid, accurate development of the inventories.

6.4 SUMMARY AND CONCLUSIONS

The previous sections of this chapter lead to a number of important conclusions concerning tools for future emission inventory development and communication. First, the examples given here demonstrate that a number of observation and analysis techniques are currently in or beyond the development stage, and offer major opportunities for improving emission rate estimation. Most of the noted techniques provide data for determining emission rates directly, allowing their results to be adopted into the inventory structure without applying the emission factor/activity factor paradigm of Equation (2.1). While these methodologies represent various levels of maturity, all show considerable potential for future use, and many can be applied to address the weaknesses of today’s inventories discussed in Chapters 5 and 7.

Second, many (but not all) of the techniques discussed here are more amenable to constructing top-down “reality checks” of bottom-up inventory estimates, rather than themselves providing bottom-up information. While this associates a somewhat secondary nature with these techniques, they are nonetheless highly relevant for three important reasons:

- Major improvements in cost-effective source characterization can be achieved with new ambient air measurements. These measurements provide direct estimates of emission rates which complement conventional stack or source testing. The array of ambient observation techniques offers important opportunities for emission observations, especially for sources such as transportation and fugitive categories, which are otherwise difficult to characterize.
- On occasion, top-down checks such as aircraft and satellite observations have indicated major errors in previously existing bottom-up inventories. Examples in this chapter and in Chapter 7 give evidence for this finding.
- Many circumstances involving multiple small and/or fugitive sources result in significant emissions, but are difficult if not impossible to characterize using conventional bottom-up techniques. Under such circumstances those techniques normally described as “top-down” become the only practical approach for emission estimates.

Third, optimum effectiveness of many of the methods described here often lies in a combination of two or more of these techniques. Aircraft observations combined with an inverse-modeling interpretation is an example of such a combination.
Fourth, potential advances in database management techniques, practices, and facilities can be expected to substantially enhance the future access, usability, and quality of emission inventory data. Although some highly visionary advancements have been suggested for the future, many, somewhat less ambitious, improvements can be incorporated over the near term. Forming a distributed emission inventory data network is a key example of such an action.

Fifth, significant improvements are possible for conventional emission inventory analysis, including refining estimates of emission factors and activity factors. Utilizing data from enhanced ambient monitoring capability, newer measurement techniques for deriving factors, and relating such data to conventional reference test methods, are particularly challenging aspects of this effort.

Finally, and as noted in the introduction to this chapter, members of the emission inventory community should recognize that they themselves are primarily responsible for pursuing this advanced agenda. To do this, it is important to establish an active, two-way dialog between emission inventory developers and scientists involved in creating future measurement and interpretive methodologies. Active promotion of this interaction is a strong recommendation of this Assessment.

Table 6.4 summarizes these techniques and provides an indication of where the different methodologies are linked with current emission inventory deficiencies. Although the tabulation does not evaluate whether new methods are superior to older techniques, it clearly shows that a variety of opportunities exist for enhancing the available information for inventory development.

The following recommendations derive from this chapter:

- Where appropriate resources are available, special efforts should be made to take advantage of the contemporary measurement methods and source-characterization experiments noted in this chapter to minimize the use of AP-42–based emission estimates, which involve “default” characterizations of undetermined local applicability.

- Increased effort should be made to maximize use of ambient measurements for deducing emission rates and chemical-source profiles, as well as for cross-checking existing emission inventories. The use of receptor-modeling and and inverse-modeling techniques, in combination with regulatory air quality modeling analysis should be encouraged for this purpose.

- Increased efforts should be fostered to increase the accessibility, transparency and timeliness of emission inventory publication by taking advantage of contemporary information technologies.

- Establishment of a continuing dialog between emission inventory developers and scientists involved in creating future measurement and interpretive methodologies will facilitate future development and the widespread application of emerging technologies exemplified here.
<table>
<thead>
<tr>
<th>Identified uncertainty</th>
<th>Satellite/aircraft remote sensing</th>
<th>Ground remote sensing</th>
<th>Alternative mobile measurements</th>
<th>Dilution tunnel</th>
<th>CEMS</th>
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<th>“Top down” modeling</th>
<th>Advanced data management</th>
<th>Comments</th>
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<tbody>
<tr>
<td>All sources</td>
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<tr>
<td>Quantify uncertainties</td>
<td>--</td>
<td>Opportunities for stationary mobile</td>
<td>Options to enhance real world data</td>
<td>Improve source data, account for semivolatiles and range of emissions</td>
<td>Large potential for stationary sources</td>
<td>Cross-check for emission factors and emission reports</td>
<td>Important for fugitives and natural emissions</td>
<td>Assist in quantifying uncertainty</td>
<td>Include uncertainty in data base</td>
<td>Special measurements limited by short-term samples/models</td>
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<tr>
<td>Small sample limitation</td>
<td>Helpful for location, and possibly large scale</td>
<td>Potentially important for stationary and mobile</td>
<td>Add substantial fleet data</td>
<td>Addresses small sample limit</td>
<td>--</td>
<td>--</td>
<td>Means for extrapolation to many sources</td>
<td>--</td>
<td>Remote sensing potential large but detection limit a problem</td>
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<tr>
<td>Spatial/temporal disaggregation</td>
<td>Assist for large scale</td>
<td>Can clarify disaggregation</td>
<td>Mostly small scale observations</td>
<td>Addresses this issue at single source level</td>
<td>Assists in spatial resolution of plumes</td>
<td>Small scale fluxes point measured</td>
<td>Means for addressing multiscale</td>
<td>--</td>
<td>Rely heavily on modeling with measurement support - Satellite or spatial/temporal measurements potentially useful</td>
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<td>Multiscale resolution</td>
<td>Assist for large scale</td>
<td>Best for small scale resolution</td>
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<td>Assists in spatial resolution</td>
<td>Small scale fluxes</td>
<td>Means for extrapolating to multiscale</td>
<td>Represent multiscale in data base</td>
<td>Modeling key to integrating point measures - remote sensing may assist</td>
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<tr>
<td>Timely update and report</td>
<td>Adopt satellite technology where possible to add to timely data access</td>
<td>--</td>
<td>--</td>
<td>Automated data shortens report time</td>
<td>--</td>
<td>--</td>
<td>Key to timely reporting</td>
<td>--</td>
<td>Organization of data mgmt. combined with continuous measurements with automation will resolve this weak</td>
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<td>Rigorous QC/QA application</td>
<td>--</td>
<td>Offers a means of cross-checking estimates</td>
<td>Cross-check on model estimates</td>
<td>Cross-check on emissions</td>
<td>Easier to formalize process</td>
<td>Adds method for cross-checking</td>
<td>Cross-check on emission models</td>
<td>Important for QC/QA</td>
<td>Add measurement cross-checks and modeling as key tools</td>
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<tr>
<td>Identified uncertainty</td>
<td>Satellite/aircraft remote sensing</td>
<td>Ground remote sensing</td>
<td>Alternative mobile measurements</td>
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<td>Documentation inadequate</td>
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<td>--</td>
<td>Formalizes reporting</td>
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<td>Important to documentation</td>
<td>Increased accessibility and transparency will depend on documents</td>
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<tr>
<td>Biogenic component not characterized</td>
<td>Potential application of land use and biogenic inference</td>
<td>Potential for small scale flux measurements</td>
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<td>Potential for flux measure</td>
<td>Important for biogenics</td>
<td>Cross-check on emission models</td>
<td>Must include reporting of biogenics</td>
<td>Increasing importance for criteria pollutants and large scale phenomena</td>
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<tr>
<td>Large uncertainty in toxic air pollutants</td>
<td>Sensors limited but potential</td>
<td>Opportunities for special pollutants, ultrafines, and HAPs</td>
<td>Capable of toxic air pollutant testing</td>
<td>Limited sensors for toxic air pollutants</td>
<td>--</td>
<td>--</td>
<td>Cross-check on emission estimates</td>
<td>Attention to data base important for improving emission inventories</td>
<td>Special concerns for measurements or sample-detection limits</td>
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<tr>
<td>Stationary sources</td>
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<tr>
<td>Uncertainty w/o CEMS</td>
<td>Qualitative locator for plumes</td>
<td>Complement CEMS for testing</td>
<td>--</td>
<td>Complement sampling for exotic species</td>
<td>--</td>
<td>Adds means of cross-check of default emission and activity factors</td>
<td>--</td>
<td>Cross-check on emission estimates</td>
<td>Need for integrating CEMS with conventional data</td>
<td>Develop low cost CEMS for deployment to new categories of sources</td>
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<tr>
<td>Multiple source averages and short sample</td>
<td>--</td>
<td>Study a subset for limits of averages and sampling</td>
<td>--</td>
<td>Facilitates multiple source averaging</td>
<td>--</td>
<td>--</td>
<td>Cross-check on emission estimates</td>
<td>Add document for limitations and uncertainty</td>
<td>Long term study use CEMS</td>
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<tr>
<td>Area/mobile sources</td>
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<tr>
<td>Limitations for fugitive and mobile emissions</td>
<td>Locator for multiscale dust storms, fires, land use</td>
<td>Potential for some fugitive/mobile source testing</td>
<td>Cross-check on model estimates</td>
<td>Important for SVOC from combustion</td>
<td>--</td>
<td>Potential for improving flux measures</td>
<td>Opportunity to reduce area source uncertainty</td>
<td>Cross-check on estimates for multiscales</td>
<td>Document limitations and uncertainty - links with models</td>
<td>Recognize increasing importance of fugitive emissions and other source types</td>
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<tr>
<td>Identified uncertainty</td>
<td>Satellite/aircraft remote sensing</td>
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<tr>
<td>Limitations from updating</td>
<td>--</td>
<td>Can add tools for update data</td>
<td>Can add observations for updating with real-world fleet</td>
<td>Provide insight on SVOC and ultratines</td>
<td>--</td>
<td>--</td>
<td>Provides a means for update fugitives</td>
<td>Document changes, upgrades and flag new data</td>
<td>Important function for data management</td>
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<tr>
<td>Mobile/ fugitive not rated for quality</td>
<td>Cross-check on location of impact from multi scale events</td>
<td>Can add data for quality check</td>
<td>Support for quality rating</td>
<td>Revise report for emissions</td>
<td>--</td>
<td>Adds a means of cross-check</td>
<td>Improves quality estimate</td>
<td>Crosscheck on multiscale estimates</td>
<td>Add documentation in system</td>
<td>Important to account for fugitive emissions and mobile fleet categories</td>
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<tr>
<td>Large uncertainties for all vehicle classes undocumented</td>
<td>--</td>
<td>Cross road measurements proven for data</td>
<td>can add observations for improving documented emissions</td>
<td>Add data for testing in laboratories</td>
<td>--</td>
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<td>Cross-check on vehicle fleets</td>
<td>Add to data management system with new data and uncertainty estimates</td>
<td>Priority steps to reduce uncertainty in fleet descriptions important</td>
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<tr>
<td>Models do not account for real world variability</td>
<td>--</td>
<td>Should enhance inventory using receptor and inverse model</td>
<td>Improve knowledge of real world fleet emissions</td>
<td>--</td>
<td>--</td>
<td>Improve fugitive and biogenic models with actual observations</td>
<td>Models needed to compare real world data with emission models</td>
<td>Facilitate linkage between data and models</td>
<td>Importance of mobile sources and natural emissions for OC increasing</td>
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<tr>
<td>Nonroad emissions based on default estimates/ uncertainty undocumented</td>
<td>--</td>
<td>Observations should improve estimating emissions</td>
<td>--</td>
<td>Use for estimation of uncertainties comparable with mobile</td>
<td>--</td>
<td>--</td>
<td>Add for cross-check</td>
<td>Add emiss. modeling link with data management</td>
<td>Need to add data to account for changing fuels</td>
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<tr>
<td>Nonroad emissions not tied to real world measurements</td>
<td>--</td>
<td>Obs. Should improve with use of remote sensing</td>
<td>--</td>
<td>Use for estimation of OC and BC, toxic air pollutants</td>
<td>Potential for inexpensive CEMS-like sensors</td>
<td>--</td>
<td>--</td>
<td>Improve estimates using adv. NON-ROAD and cross-check measures</td>
<td>Add to data management platform with link to models</td>
<td>Need to add data for alternate fuels</td>
</tr>
</tbody>
</table>
REFERENCES FOR CHAPTER 6


CHAPTER 6


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Pierson, W., Brachaczek, W. 1983. Particulate matter associated with vehicles on the road. II. Aerosol Science and Technology 2, 1–40


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CHAPTER 6


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