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Chapter three: methodology of exposure modeling

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Abstract

In this chapter, the concept of exposure assessment and its evolution is introduced, and evaluated by critically appraising the pertinent literature as it applies to exposures to Particulate Matter (PM). Exposure measurement or estimation methodologies and models are reviewed.

Three exposure/measurement methodologies are assessed. Estimation methods focus on source evaluation and attribution, sources include those outdoors and indoors as well as in occupational and in-transit environments. Fate and transport models and their inputs are addressed to estimate concentrations outdoors and indoors; source attribution techniques help focus on the contributing sources. Activity pattern techniques are also reviewed and their use in exposure models to estimate inhalation exposure to PM is presented. Deterministic, regression and other stochastic models of exposure to PM are reviewed and evaluated.

Strengths, limitations, assumptions and affirmations of the use of exposure assessment as an integral component of risk assessment and risk management are discussed in the conclusions and discussions section of this work.

Keywords: Exposure models; Source inventories; Meteorology models; Indoor air quality models; Activity patterns; Time budgets

Contents

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1. Introduction

Though used widely by many disciplines, exposure assessment is a relatively new scientific endeavor of public health. Its inclusion as an explicit component of risk assessment started in the 1970s. The evolution of risk assessment from the conventional six-step process to the present seven-step methodology is illustrated in Fig. 1 (Moschandreas and Saksena, paper in this issue).

Because the term "exposure" is used to denote different concepts, the use and comparison of exposure study results can be difficult. In this chapter, exposure denotes the contact between an agent and the boundary of a receptor. An agent is a substance known or suspected to be toxic to the receptor. Receptors can be humans, animals, trees, historical buildings or any other ecological entity. The boundary of contact is an external boundary. This chapter limits itself to one agent, the class of particulate matter (PM) suspended in the atmosphere, to human receptors, and to the nose/mouth inhalation boundary. PM is further classified by mass concentrations in the total suspended particulate (TSP), PM_{10}, and $PM_{2.5}$ size fractions (particles with aerodynamic diameters less than ~30, 10, and 2.5 μm, respectively).

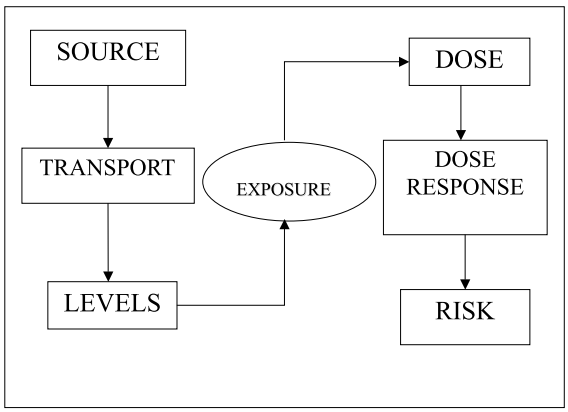

Fig. 1. Risk assessment process: The early version of the risk assessment process (in rectangles) did not include the exposure element.

Although inhalation may not be the most important pathway of exposure for all pollutants, it is considered the one of major concern for exposure to PM. Related concepts, such as dose, will not be addressed in this chapter.

The National Academy of Sciences suggests the following model as the fundamental expression for estimating inhalation exposure to an agent (NAS, 1991):

$$E = \sum C_{ijk} t_{jk} \tag{1}$$

where E is the inhalation exposure, the output of the modeling effort, C_{ijk} is the concentration of agent i, assumed to be constant time, t_{jk} in microenvironment (μE) j where subject k spends time, and t_{jk} is the time spent by subject k in microenvironment j.

The following, more detailed, generic exposure model should be considered in studying inhalation exposure to PM and its constituents; it estimates total exposure as a sum of exposures at each of the microenvironments:

$$E_{ijk} = \sum_{ik}^{\text{outdoors}} C_{ik} t_{ik} + \sum_{ik}^{\text{indoors}} C_{ik} t_{ik} + \sum_{ik}^{\text{occupational}} C_{ik} t_{ik} + \sum_{ik}^{\text{in-transit}} C_{ik} t_{ik} \tag{2}$$

The symbols and subscripts denote variables indicated in Eq. (1). Eq. (2) specifies the four microenvironments that must be addressed; other microenvironments are available and could be included but are considered of less importance in this methodology chapter.

If the concentration were not assumed constant over the time period of interest, then the above equation in-

volves integration over time. Other exposure expressions are used to estimate exposures to pollutants in the ingestion and dermal absorption pathways.

Major variables of concern in the estimation of exposure, Eq. (2), are the concentration of PM and its constituents outdoors, in the residential environment, in the work place and in-transit, and the time each subject spends in each microenvironment. Investigators may measure these concentrations or they may estimate them. Relevant variables for estimating exposures are:

- major sources of PM in each of the microenvironments and their emission rates,
- meteorological conditions that affect the transport of pollutants from their source to the receptor site,
- other variables in fate and transport models selected by the investigator for estimating agent concentrations outdoors,
- several indoor variables that affect the amount of agent that infiltrates indoors,
- time that subjects spend in each microenvironment.

The exposure scenario includes a list of all sources considered and their emission rates, activity patterns of receptors, and their time budgets, demographic characteristics of the subjects, descriptions of microenvironments and other attributes that describe fully the exposure setting that is estimated by the investigator. Ideally, the output of an exposure assessment is the exposure profile that denotes the magnitude and the spatial and temporal variation of exposure.

The expression of exposure as the sum of exposure through all pathways (inhalation; ingestion in food, water, or by putting fingers in the mouth; absorption through the skin, etc.) and in all microenvironments in which the subject spends time (outdoors, at home, at work, during transit, etc.) has proven to be a useful concept for understanding exposure to single chemical species, such as lead, or to classes of chemical compounds, such as chlorinated hydrocarbons from water purification. PM is a complex mixture of particles of different sizes, having different chemical and physical properties, being produced by different sources, and having different types and degrees of toxicity.

An alternate expression for inhalation exposure to PM has been suggested by Wilson and Mage (1999). Instead of the sum of exposure to all particles in several microenvironments, exposure is expressed as the sum of exposure in all microenvironments to particles from several sources. Thus, instead of expressing exposure as the sum of exposure outdoors, at home, in-transit, and at work, exposure is expressed as the sum of exposure to particles of ambient origin (both ambient particles while outdoors and ambient particles that have infiltrated indoors while indoors), particles of indoor origin, particles due to occupational activities, and personal activity particles (particles due to smoking, hobbies or other activities that impact the subject but not other people in the same microenvironment).

This expression of inhalation exposure to particles can be readily used if the concentration of particles of ambient origin, that have infiltrated into an indoor microenvironment, is calculated from the ambient concentration, the properties of the particles, and the properties of the microenvironment. However, if the model is based on measured concentrations of particles indoors and outdoors, it will be necessary to disaggregate the total PM found indoors into particles of ambient origin that have infiltrated indoors from outdoors, particles of indoor origin, and particles due to personal activity.

The expression of exposure to PM in terms of particles from different sources is useful for several reasons. First, it is useful for risk assessment and risk management. Currently, some information is available on the relative toxicity of combustion products from various types of fuels when used in open, unvented combustion indoors. Relationships have also been found between ambient particle concentrations and a variety of health outcomes. No exposure–effect relationships have been determined for particles. Toxicological studies suggest that particles of different sizes and from different sources will have effects that differ in both type and degree. This is also suggested by the literature on uncontrolled indoor combustion. If this concept of differential toxicology is accepted, knowing exposure as a function of source will allow targeting exposure reduction to the most significant sources. In developed countries, fine particles of ambient origin are probably of most concern. In homes where fuels other than electricity are used without adequate ventilation, particles of indoor combustion origin may be of most concern. Relating health outcomes to sources allows more effective risk management by concentrating control on the most dangerous sources.

This recent alternative to the conventional exposure measuring/estimating methods, presented in June 1999 (Wilson and Mage, 1999), articulates a new avenue for associating exposures to PM with sources and health effects. The traditional methodology is to use one or more of three methods for measuring or estimating exposure: the direct, indirect or reconstructrive methods (NAS, 1991).

The direct method *measures* the exposure of individuals to a pollutant(s) of interest using small and unobtrusive exposure sampling devices. This method is also known as the point-of-contact method and measures the concentration of the pollutant at the point or close to the point of contact with the individual. This technique requires the cooperation of the subjects participating in the exposure measurement (US EPA, 1997).

The indirect method *estimates* exposures by measuring or estimating pollutant concentrations at different microenvironments where people spend their time. These concentrations are weighted by the amount of time individuals spend in these locations. This approach, known as the scenario approach, requires measured or estimated pollutant concentration, time-of-contact, demographic and time budgets and activity information of the exposed individuals. Building characteristics that affect infiltration and indoor emissions, and inventories of potentially contributing outdoor and indoor pollutant sources are often obtained from questionnaires. Outdoor concentrations of pollutant(s) may be measured at community representative sites or estimated from source information combined with fate and transport models. Inhalation rate can also be estimated from activity information.

The reconstructive method *measures* levels of either the agent itself or its metabolite in a target organ, and *estimates* exposure levels backwards using pharmacokinetic models. It is possible to estimate exposure through measurement of a biological marker. There is a continuum between biological markers of exposure and of effect. At least theoretically, if the mathematical relation between the various points on this continuum is fully understood, then an outcome measure can be used to reconstruct the extent of exposure. In fact, this is difficult and is limited thus far to just a few examples. The difficulties are caused both by inherent variations in human biology and by uncertainties in the pathogenic processes between exposure and effect. Biological markers on the exposure side of the continuum provide a closer linkage to external exposure. This is particularly true for measures of blood or tissue levels of the compound itself, e.g., blood lead, exhaled benzene, and of its ligands, e.g., carboxyhemoglobin.

Linkage of biological markers to external exposure requires understanding of the four basic components of absorption, distribution, metabolism and excretion. Physiological based pharmacokinetic (PBPK) models are used to estimate the relation between these different compartments and to the external exposure levels.

Recent advances in analytical techniques, and particularly in the field of molecular biology, provide the opportunity to develop new biological markers of exposure and to improve our understanding of the linkages between external exposures and biological markers of the continuum between exposure and effect.

The focus of this chapter is on the methodology of the direct and indirect methods of exposure estimation, thus the reconstructive or other alternative methods are not discussed any further. The objectives of this chapter are to:

1. Summarize methodologies for estimating or measuring inhalation exposure to PM and its constituents.
2. Rank methods used in the literature as a function of intended use of the results, complexity, accuracy and resources.

Section 2 of this chapter addresses inputs and outputs of the indirect method for assessing exposure, Section 3 reviews the direct method, while Section 4 addresses advantages and shortcomings of methods discussed and provides recommendations.

2. The indirect method of exposure estimation and source attribution

The indirect method of exposure estimation models or measures agent concentrations and the time people spend in each microenvironment. These inputs are then used in Eq. (2) to estimate exposures (see Fig. 2). The microenvironmental concentrations can be further attributed to indoor and outdoor particle emissions by source or re-

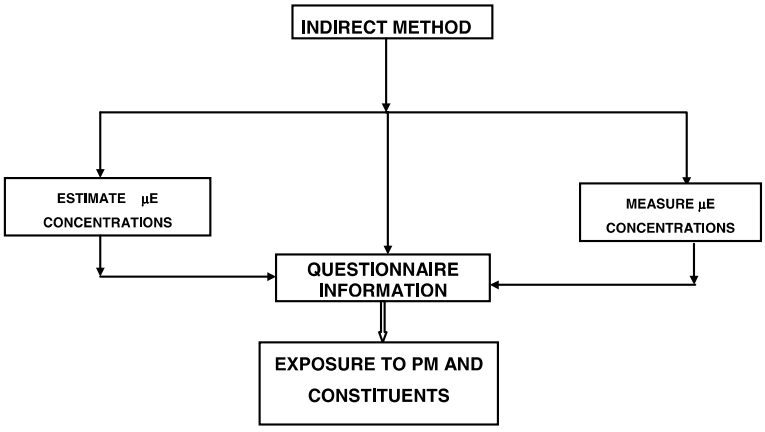

Fig. 2. Options and components of the indirect method of calculating exposure.

ceptor modeling. Source models estimate receptor concentrations from source emissions and meteorological measurements and do not require microenvironmental sampling except for model validation. Such validation is usually necessary for at least some of the estimated concentrations so it cannot be completely eliminated. Receptor models infer contributions from different source types by measuring chemical and physical properties of the sampled particles that are indicative of their origins. These samples can be taken at fixed indoor or outdoor monitors or mobile samplers for the indirect method, or from monitors that follow the activities of representative individuals through their daily activities. The application of both source and receptor models allows for further validation of both models, thereby increasing confidence in their source attribution results.

2.1. Source model estimates of concentrations and source contributions

Source modeling consists of three components: (1) emissions rates from sources as a function of time and location; (2) wind speed, wind direction, temperature, relative humidity, pressure, and other atmospheric properties at different times and locations that affect the transport, diffusion, and deposition of the emitted pollutants; and (3) transport, diffusion, deposition, and chemical transformation mechanisms that simulate changes in emissions between source and receptor.

2.1.1. Emissions rates and composition

Emissions inventories document temporal and spatial emission rates based on activity levels, emission rates per unit of activity, and meteorology (US EPA, 1998). Emission rates per unit activity, or *emission factors*, are determined from tests of representative sources within a category. Representative industrial sources are usually tested by hot stack sampling, in which air is extracted and passed through a filter that is weighed before and after sampling to measure particle loadings. Motor vehicle exhaust emissions are sampled from diluted exhaust streams when on-road emissions are simulated on a laboratory dynamometer. Residential fuel combustion for heating and cooking emissions are also simulated in laboratory conditions. Fugitive dust emissions from paved and unpaved roads, construction, and agricultural operations are simulated estimated from upwind and downwind sampling.

Emissions factors derived from many tests on outdoor sources in the US are compiled by US EPA (1999) and are regularly updated. Emissions factors for particles from indoor sources are not compiled in a single resource and must be assembled from individual studies (e.g., Yocom, 1982; Lionel et al., 1986; Koutrakis et al., 1987; Bare, 1988; Lewis, 1989, 1991; Lofroth et al., 1991; Samet and Spengler, 1991; Yocom and McCarthy, 1991; Kim and Fergusson, 1993; Raiyani et al., 1993; Schare and Smith, 1995; Wallace, 1996; Lioy et al., 1999). The emissions factors derived for one source type or area may not completely represent actual emissions from similar source types in other areas. Emissions factors derived for the United States almost certainly do not represent the emitters in other countries, although they are often used as a first approximation.

Emission factors are applied to activity levels that often can be determined from databases compiled for other purposes in a study area. Commonly used activity levels are: (1) fuel consumed, product produced, or material inputs for industrial sources; (2) vehicle miles traveled, fuel sales, miles of roadway, or vehicle registrations for mobile source emissions; and (3) population density, number of households or businesses, amount of residential wood or coal sold for area source emissions. These data are often available from census statistics, traffic demand models, roadway maps, tax records, land use maps, and economic surveys.

For multi-component pollutants such as PM and volatile organic compounds (VOC), source profiles are used to allocate total mass emissions to chemical components. Source profiles are determined by taking samples from representative sources and submitting them to laboratory analysis for the desired chemical species. Source profiles are also determined from representative tests and represent the fraction of total PM or VOC mass composed of a specific chemical species for more details on this subject see article in this issue by Watson and his associates, which describes the organic and inorganic components found in various source emissions and summarizes compilations of the profiles, mostly from US sources.

Activity data are often unavailable for the spatial scales desired of source models. US EPA (1998) annual emissions estimates for the entire United States have the resolution of US counties. This resolution is usually insufficient for source modeling. Emissions rates are often estimated for a larger area (e.g., a county, state or province) and spatially allocated to a smaller area based on a population census, land use map, or roadway network. An accurate inventory for source modeling often requires greater effort and expense than field monitoring for concentrations and meteorology.

Emissions inventories are often used as source models without meteorological measurements to develop control strategies for linear rollback (Barth, 1970; deNevers and Morris, 1975; Cass, 1981; Cass and McRae, 1981, 1983). Rollback assumes that atmospheric concentrations in excess of background are proportional to aggregate emission rates. Reducing excessive concentrations of a pollutant to levels below a preset standard requires emission reductions that are proportionally equal to the relative amount by which the standard is exceeded.

Common categories of outdoor particle emissions sources that must be considered in outdoor air are:

Gasoline exhaust: Every urban area includes vehicle exhaust from light duty cars, light to medium duty trucks and buses, and small engines. Emissions inventories do not usually contain information about cold-starts and visibly smoking vehicles, although these might be discriminated by certain organic compounds in a profile. Leaded gasoline is no longer used in the US, but it is common in many countries and results in larger particle emissions because vehicles that use it do not have catalytic converters. Two stroke engines in small vehicles, motorcycles, and utility engines combine oil with gasoline and have substantially higher particle emissions per quantity of fuel consumed compared to four-stroke engines. Fuel sales and vehicle miles traveled are the best ways to estimate the magnitude of emissions. These can be spatially allocated based on vehicle counts from different types of roadways and roadway network maps.

Diesel exhaust: Every populated area contains heavy-duty trucks and buses with diesel engines. Some areas may also contain off-road equipment, stationary engines for pumps and generators, and locomotives. Fuel sales and vehicle counts with spatial allocation by roadway are used to determine emissions in space and time.

Residential cooking, heating, and burning: Cooking is common in all populated areas, while heating and burning depend on climate and local regulations. The appliance type and fuel make the biggest difference in emissions rates. Natural gas, oil, coal, wood, trash, and other biological materials are often consumed in these activities. Differences in the types of appliance used and burning practices also cause large variations in emissions factors. Numbers of residences and fuel sales are often used to determine the magnitude of these emissions and to allocate them to their locations.

Fugitive dust: Fugitive dust results from non-ducted open areas such as paved roads, unpaved roads, agricultural tilling, construction, wind erosion, and industrial aggregate. Cement, quarrying, and mining are the main industrial emitters. Miles of roadway, locations of construction sites, and maps of agricultural fields and open areas are used as the bases for estimating the magnitude and locations of emissions.

Industrial point sources: Power stations, copper smelters, lead smelters, steel mills, aluminum mills, and other large industries are easily identifiable and have fixed locations. Their non-fugitive emissions are typically ducted through stacks and emissions factors can be measured. Fuel use and amount of product are usually used to estimate the magnitude of emissions.

Indoor emissions inventories include the following particulate source types.

Infiltrated outdoor air: Doors, windows, and walls can be treated as emitters, typically through the application of an infiltration model. These depend on an accurate estimate of outdoor concentrations, air transfer rates, and particle losses as they pass through narrow openings, especially when doors and windows are closed.

House dust: House dust results from infiltration, track in, and indoor generation activities. It is often suspended by vacuuming and sweeping as well as indoor personal movement.

Cooking and heating: Fuel combustion as well as the type of food being cooked and the cooking method cause indoor emissions. A large fraction of these may be vented by a cooking hood with an exhaust fan to outdoors.

Environmental tobacco smoke (ETS): Cigarettes, cigars, and pipes create directly emitted and inhaled and exhaled particles. These can linger and disperse throughout an enclosed area.

Animal dander and fur: Domestic pets such as dogs and cats shed skin and hair. Human occupants also represent a source of these.

Molds, spores, and fungi: These material forms in moist locations and in house plants. Ventilation ducts are often breeding grounds as well as methods to circulate these throughout an enclosure.

Vehicle exhaust: Although externally emitted, a fraction of vehicle exhaust enters into the passenger area. This is also an important infiltration source during commuting as most vehicle compartments are close to the exhaust from neighboring vehicles. Warming up cars in indoor garages creates exhaust that can infiltrate into living areas of a home.

Occupational emissions: There are a large variety of occupational emissions, depending on the type of work involved.

2.1.2. Meteorological data

Meteorological data are a fundamental inputs for the accurate assessment of the fate and transport of air pollution through modeling. These data are used to either calculate the concentration of a pollutant at a receptor through dispersion, as in the case of source models, or to determine possible source regions contributing to measured concentrations at a specific receptor (receptor models). Consequently, all models that determine fate, transport, or origin of atmospheric pollutants depend on meteorological parameters as input.

All dispersion models require core parameters (Table 1). These include surface wind direction, wind speed, air temperature and cloud data. In addition, upper air data, typically measured twice per day, determines wind, temperature, and humidity changes with height (Schulze and Turner, 1996).

Steady-state Gaussian plume dispersion models require hourly single-point meteorological data at the surface and an upper air station to estimate the mixing

Table 1
Typical meteorological data requirements and characteristics for steady-state and non-steady-state dispersion models

	Steady-state models	Non-steady-state models
Data characteristics	Discrete data One-dimensional data Fine temporal resolution	Gridded data Three-dimensional data Fine temporal resolution
Core parameters	Wind direction Wind velocity Temperature Upper air data	Wind direction Wind velocity Temperature Upper air data
Additional parameters		Precipitation Relative humidity Turbulence parameters Atmospheric pressure Solar radiation Sea surface temperature

height. More sophisticated models, such as non-steady-state puff and Eulerian models generally require more comprehensive meteorological data from multiple meteorological stations (Table 2). In addition to the core parameters, these models require atmospheric pressure, humidity, precipitation, turbulence parameters, solar radiation, and where applicable, sea-surface temperature data.

The representativeness or adequacy of meteorological data is an important element in accurately modeling the dispersion of air pollution. The observational meteorological data used for pollution modeling is generally regarded as adequate if a sufficiently dense coverage of stations at the surface and aloft exists over the modeling domain, including all relevant physical and climate zones. In addition, all the meteorological parameters necessary for modeling have to be measured on a fine temporal resolution. Often, especially in developing countries, the spatial coverage and temporal resolution of observational surface and upper air meteorological stations is inadequate, or even non-existent over the domain where dispersion modeling is to take place. Accuracy of the data can also be questioned where inadequate instrumentation, training and personnel render data subjective.

A common error made by investigators when observational data is unavailable in the modeling domain is to use data from distant stations such as airports. These distant measurements often do not adequately represent conditions in the study area owing to differences in terrain, coastal effects, urbanization, and synoptic features.

Local terrain generally modifies wind conditions through a number of processes. Firstly, by channeling wind along valleys or around topographic obstacles, thereby resulting in directional and/or velocity modification. Secondly, local topography such as katabatic/mountain flow by night and anabatic/valley flow by day create a unique, localized circulation that can return aged pollutants to mix with fresh emissions.

Land–sea temperature differences result in a thermally driven circulation. By day, a sea breeze with onshore flow at the surface may develop that progressively extends inland by late afternoon to up to a 100 km or more in some cases. A return flow aloft is often associated with the sea breeze. At night, the reverse occurs with the establishment of a land breeze with offshore flow at the surface. This diurnal circulation may have important ramifications for the dispersion of atmospheric pollutants and the use of distant observational data may not reflect such circulation.

Modification of the local energy budget through urbanization has the effect of locally increasing atmospheric turbulence and mixing height, with important consequences for the dispersion of pollutants over the city. Also, increased roughness length and urban canyons modify the urban wind characteristics significantly over relatively short distances.

The representativeness of observational data from meteorological stations distant from the modeling domain may also be affected by synoptic weather events. Specifically, the remote observational station may be influenced by transient meso- and synoptic-scale weather systems not affecting the modeling domain. This may lead to an inaccurate assessment of meteorological conditions in the study area.

Table 2
Comparison of meteorological data generation

Type	Observations				Generated NWP models
	Surface stations	Upper air stations	Mast	Remote sensing (tethersonde/SODAR)	
Vertical resolution	Single (1.2 m)	Up to ~50 mb	10–100 m	Up to 1500 m	~5–25 levels (up to 300 mb)
Horizontal resolution	Discrete data	Discrete data	Discrete data	Discrete data	Gridded data
Temporal resolution	1 hourly	12 hourly	1 hourly	1 hourly	1 hourly
Data collection period	>1 year	>1 year	>1 year	>1 year	~3–5 months

A number of solutions are possible to remedy these potential shortcomings of observational meteorological data. These include the establishment of an improved observational network representing conditions in the study area, or, in what is usually a more cost-effective option, the generation of site-specific meteorological data using meteorological models.

Site-specific meteorological data can be collected for an area with a paucity of existing data using a number of means. Firstly, a weather station, where hourly surface observations can be complemented with twice daily upper air soundings can be considered as a long term investment by weather services. Secondly, a meteorological tower with continuous measurements for heights up to 100 m. Thirdly, measurement of wind velocity, direction, temperature and humidity profiles within the mixing layer can be conducted through tethersonde or remote sensing devices.

Monitoring is generally time-consuming and expensive. All monitoring methods take longer than a year to collect a year of data; and operating costs are high. For example, the costs for a single meteorological tower for one year are typically in the range of several tens of thousands of dollars for measurements at a single location. Data coverage over the domain may still be inadequate with a single measurement station, so often multiple measurements are required to adequately represent meteorological conditions.

An alternative method is to apply prognostic numerical weather prediction (NWP) models to establish the meteorological conditions where few or no observational data exist. The model output is typically gridded on a fine resolution (typically 10–30 km horizontal resolution), and at 15–25 levels in the vertical. Wind speed, wind direction, temperature, humidity and other variables are produced by the model output to cover the domain. In a data assimilation mode, the NWP models use whatever observational data may be available to "nudge" the solutions of the relevant equations toward the observations. The NWP data can then be combined with fine-scale geophysical features to produce fine-scale, site-specific localized meteorological fields.

NWP models can be used to generate meteorological fields relatively quickly. When run in a data assimilation mode, it would typically require 3–6 months to generate a full year of meteorological data up to the size of an entire medium-sized country for approximately the same cost as a single monitoring site. Data from such a modeling study can be generated for any part of the world using routinely available global model data sets. Once generated, this synthetically generated meteorological data set can be used for many years for different modeling studies within the domain.

An example of this approach to acquisition of meteorological data is a study conducted in western Canada. An irregular network of widely spaced surface and upper air stations in British Columbia and Alberta resulted in a paucity of data coverage over much of these large provinces. The Mesoscale Model Version 5 (MM5) model (Grell et al., 1994) was used to generate a full-year database of hourly meteorological profiles at 20 km resolution at over 8000 locations. For about the cost of constructing and operating a single meteorological tower, the use of a meteorological model generated an annual data set of hourly meteorological profiles covering the entire two-province area. In addition, the NWP generated meteorological fields are of a finer temporal resolution than the observed upper air data (hourly as opposed to twice daily). The US EPA sponsored the generation of a similar database covering the continental United States, southern Canada, and northern Mexico with 80 km horizontal resolution. This meteorological modeling approach is especially suitable for cost-effectively generating meteorological databases in developing countries where observational networks may be even less fully developed than in Canada and the United States.

2.1.3. Fate and transport models

For evaluating the inhalation component of exposure, source models are used to produce estimates of pollutant concentrations in the ambient air at receptor points placed within the modeling domain. As shown in Table 3, the model requires information on the location, strength, and type of various sources, pollutant properties, meteorological data, and geophysical characteristics of the area to estimate pollutant transport, dispersion, chemical transformation, and depletion. The results are a set of pollutant concentrations at arbitrarily defined points (often at thousands of points throughout the modeling domain) for each pollutant of interest. The modeling produces time series of hourly average concentrations at each receptor point for each pollutant. Coupled with population census data and time budget information for categories of individuals, the outdoor exposure to each pollutant can be estimated. The ambient concentration predictions of the source model may be used as input to other models such as indoor air pollution models and in the occupational and in-transit exposure assessment.

In general, source modeling is applied when there is a reasonable understanding of the emissions inventory. In some cases, primarily for major point sources such as power plants, chemical plants, refineries, or incinerators, the emissions data are based on direct measurements made at the stack. More often, the emission estimates come from application of empirical relationships based on similar types of sources, from surrogates such as population density or fuel consumption, or from emissions models (e.g., the MOBILE5.0 model for traffic emissions, or the emissions production model (EPM) (US Forest Service emission model from fires)).

Table 3
Fate and transport models—input requirements

Input requirement	Description	Source of data
Pollutant source inventory	• Emission rates (including diurnal, seasonal or arbitrary variation factors)	• Previous inventories
• Point sources	• Stack parameters	• Surrogate measures (population, fuel use, etc.)
• Mobile sources	• Source locations	
• Area sources		• Empirical emission factors (e.g., AP-42)
		• Direct measurements (source testing, CEM measurements)
Pollutant properties	• Chemical characteristics (pollutant solubility, reactivity, etc.)	• Chemical databases
	• Particle size distribution	• Empirical reference data
		• Direct measurements
Geophysical data	• Terrain elevations	• Worldwide terrain and landuse datasets
	• Land use categories	• Buoy/ship datasets
	• Overwater data (air–sea temperatures)	• Model output of derived parameters
Meteorological data	• Wind speed, direction	• Surface observations
	• Temperature profiles	• Upper air observations
	• Humidity	• NWP model output
	• Pressure	• Satellite data
	• Cloudiness	
	• Precipitation	

The inventory of emissions is an important input into the source model. The modeling is best performed iteratively with feedback from ambient concentration observations or results from receptor modeling used to correct or refine elements of the inventory. Although it is *essential* to have a good quality meteorological database to drive the fate and transport model, it is possible to derive the necessary meteorological data from the output of a prognostic NWP model (see Section 2.1.2), even in the absence of local observational meteorological data. Diagnostic meteorological models can be used to develop site-specific, fine-scale wind flow fields reflecting the local transport conditions using either the NWP model output and/or local observational data as input.

The other data sets required by the modeling (pollutant properties and geophysical data) are normally obtained from *standard databases*. The exception is information on local particle size distributions, which should be derived from local data.

Source modeling estimates source contribution from individual emitters at many receptor points over a wide geographical area including pollution monitoring sites. The influence of factors such as terrain elevation, the presence of water bodies, urbanization and variations in surface conditions can be evaluated. Air quality measurements have limited and often undefined zones of representation due to these factors. Modeling can help better define a monitoring station's zone of representation. An air quality model can provide quantitative information about individual or group source contributions and detailed time- and space-resolved concentration patterns. Air quality model results assist in deciding source siting, land use planning and effectiveness of proposed emission reduction strategies. Models can develop real time, short term meteorological and dispersion forecasts leading to the possibility of operational emission controls to mitigate impacts during high exposure episodes.

An advantage of source modeling for developing countries is that modeling costs are low when compared to monitoring programs. Simulation of one or multiple year periods can be done with the simpler plume and puff models in the time frame of months (once emissions estimates have been established), with fairly modest (PC based) computer resources. The accuracy and precision of source model estimations under local conditions should always be evaluated by independent means.

2.1.4. Overview of source modeling approaches

Table 4 summarizes the major strengths and weaknesses of each of the four modeling approaches: steady-state plume models, non-steady-state puff models, particle models, and Eulerian grid models. More details are provided in the article by Scire and D'Abreton (paper in this issue). The most widely used type of air quality model is the steady-state Gaussian plume model (Turner, 1970, 1979) as implemented in the industrial source complex short term (ISCST3) model (US EPA, 1995), and in many other community codes.

The Gaussian plume model provides relatively robust concentration estimates when the basic assumptions of

Table 4
Summary of model features

Type	Examples	Advantages	Limitations	Computational requirements
Steady-state (plume) models	ISCST3 FDM	• Simplicity • Moderate data requirements • Modest computational requirements • Simulate annual or multi-annual periods	• Instantaneous transport • Straight line plumes • No pollution build-up • Uniform meteorological conditions • Calm wind conditions not modeled	Low PC
Non-steady-state (puff) models	CALPUFF INPUFF	• Realistic transport • Causality • Pollution build-up • Non-uniform meteorological conditions • Calm conditions • Linear chemistry • Source contributions • Simulate annual or multi-annual periods	• No non-linear chemistry • Puff superposition • Costly for shear and puff coherence	Low to moderate PC
Particle models	KSP MODTRAC	• Puff advantages • Shear effects • Episodic modeling only	• High level of expertise required • Limited evaluation history • Complex	High to very high workstation
Eulerian grid models	CAMx UAM	• Non-linear chemistry • Shear effects • Large sources inventories • Episodic modeling only	• Numerical diffusion • High level of modeling expertise required • Source contributions difficult	High to very high workstation or supercomputer

the model are met. However, plume models are often applied inappropriately in situations well beyond the conditions for which they are valid. In selecting a modeling approach, it is essential to understand the limitations and assumptions of the technique.

The most important assumptions of typical steady-state models are: (1) wind speed, wind direction and atmospheric stability are constant throughout the study domain; (2) plumes move in straight lines; (3) when a receptor is downwind of the source, the plume reaches the receptor, regardless of the source–receptor distance; (4) only emissions from the current time step affect the concentrations at a receptor during that time step; (5) there is always a measurable wind and calm winds are not treated.

Meteorological conditions are often approximately uniform over short distances (over distances of a few kilometers) in flat terrain. However, this is often not the case over larger distances or in complex terrain, coastal environments, or in urban areas. Wind and dispersion conditions can change dramatically in these situations and emissions from prior hours may be significant contributors to pollution levels in the current hour.

In the steady-state plume modeling approach, each hour is independent of every other hour. There is no ''memory'' of emissions released during previous time steps, and as a result, the model cannot handle situations such as stagnation, recirculation, valley trapping or nocturnal build-up of pollutants. These are common occurrences in complex terrain situations (stagnation and valley trapping) or in land–sea breeze conditions (recirculation). The assumption of steady-state conditions implies that the emission time is long compared to the transport time of the pollutant to the receptor. It also implies that the meteorological conditions (wind speed, direction, stability, etc.) do not change during the transport. This assumption is unlikely to be valid much beyond 10 km in most situations. In complex terrain applications, the assumption of straight-line plume trajectories may even be invalid a few hundred meters from a source if, for example, the valley orientation changes.

Steady-state plume models do not account for the ''causality'' effect, i.e., they assume that source emissions immediately disperse to the receptor without accounting for source/receptor transport time. This assumption is invalid for travel times that approach a typical time step of 1 h. For wind speeds of 1–5 m/s, maximum distances from a source are too short for urban scale source assessments. Steady-state plume models should not be used beyond about 10 km from a source due to causality considerations, even if the other conditions for their use are met. Although a cut-off distance of 50 km for the

steady-state models has been used in the past (US EPA, 1995), the decision was driven by the lack of appropriate modeling tools for treating causality effects. Modeling techniques have advanced sufficiently to permit application of non-steady-state models that simulate complexities of urban and regional scale.

Contributions from surface or near-surface emissions of low buoyancy pollutants are often highest during stable light/calm wind events because dilution is minimized, and vertical dispersion of the emissions is reduced. Steady-state plume models assume that the downwind dispersion is negligible relative to plume advection, which is not valid under light wind speed conditions. Steady-state models either enforce a minimum wind speed of about 1 m/s or do not perform calculations during the light wind speed events. Therefore, surface releases of non-buoyant pollutants are not properly simulated by the standard steady-state model during the meteorological conditions expected to lead to their worst case impacts.

Puff models represent emissions as a number of discrete packets of pollutant material. Each packet (or puff) is allowed to move with the local wind and evolve in time independent of other puffs. The puff approach does not require that steady-state conditions exist, so it can be applied to a larger range of conditions than a steady-state model. Two of the more commonly used puff models are the INPUFF model (Petersen, 1986) and the CALPUFF model (Scire et al., 1999). As meteorological and emission approach steady-state conditions, puff models are often constructed to approach the steady-state plume results. Thus, to the extent that the steady-state models are valid, the puff model will reproduce their results. However, when conditions deviate from steady-state, the puff model better represents pollutant transport, dispersion and accumulation.

The main advantages of the non-steady-state model are that they use full three-dimensional meteorological fields with spatial and temporal variability. Thus, changes in the flow due to terrain, water bodies, local, meso-scale, or synoptic-scale are accounted for in the simulations. Curved trajectories and causality effects (source receptor travel times) are represented. Contributions from earlier emissions, such as build-up during stagnation or recirculation are added to the contributions reaching the receptor during the current hour.

Puff models can be run with commonly available desktop computers. Simulation times are perhaps 10 times those of steady-state models for a large number of sources, but a full annual simulation can be completed in one day or one week on a PC.

Puff models require a higher level of modeling expertise than steady-state models, especially in the definition of the meteorological fields. This can be achieved by training programs. Assumptions of puff superposition, linearity of chemical reactions, lack of puff splitting during wind shear and puff coherence need to be evaluated. Some of the complexities can be addressed by appropriate selection of puff model software options.

In a particle model, the plume is represented as a series of individual particles, sometimes hundreds or thousands of particles that are individually transported through the atmosphere (e.g., the kinematic simulation particle (KSP) mode, Yamartino et al., 1996). Each particle is subjected to advection by a mean wind and to perturbations by a turbulent wind fluctuation. Particle models offer the potential to simulate both the mean concentration as well as the concentration distribution about the mean. This distribution simulates short term peak concentrations (e.g., for highly toxic acute exposure over minutes or seconds, or in odor assessments).

Particle models treat highly complex flows, including strongly sheared flows and spatially inhomogeneous meteorological conditions. Because particle models are non-steady-state models, many puff model features apply (e.g., ability to treat causality effects, pollutant build-up during stagnant conditions, and low wind speed conditions).

Particle models are computationally intensive, requiring high-end workstations or supercomputers; particle models are not currently practical for large source inventories or for long simulation times. There is a high level of expertise needed for running particle models, and substantial data requirements, including a high quality meteorological data set including turbulence fields. The operational evaluation history of particle models is limited, so it is not clear if the theoretical advantages of the approach will be realized in practice with actual data sets. At this time, particle models are primarily research models rather than operational tools.

Eulerian or grid models solve the equation of conservation mass through numerical methods. The atmosphere is divided into coupled boxes or grid cells within which pollutants are assumed to be uniformly mixed. Table 4 lists several examples of common Eulerian models. These models accommodate non-linear chemical reactions, which are necessary for photochemical modeling that estimate ozone and secondary aerosol concentrations. Simulation time is independent of the number of sources but increases with the square of the number of grids and linearly with the number of boxes. For containing thousands to tens of thousands of sources, there is a crossover point at which Eulerian models become less computationally intensive than puff or particle models. Eulerian models handle highly sheared flows well.

Disadvantages of Eulerian models include high computational costs, numerical diffusion and insufficient mass conservation (associated with the approximate numerical solutions of the conservation equations), the need for substantial computer and modeling expertise, and the treatment of each grid as a well-mixed box.

Some grid models imbed plume or puff models to account for the fact that source emissions do not mix instantaneously throughout the grid containing the source. This immediate grid mixing does not represent sub-grid-scale chemical reactions or concentrations close to an emitter. Individual source contributions are difficult to estimate with Eulerian grid models, as there is no unique solution that accounts for the individual contributions to the total concentrations at a receptor. The model must be run with and without the emissions of interest to determine individual source contribution.

2.1.5. Indoor models

Computational fluid dynamics (CFD) and Markov chain models are research methods used to estimate multi-zone indoor concentrations or three-dimensional indoor pollutant concentrations. Development of these models is in progress and their evaluation has been limited to studies performed in chambers. Both of these types of indoor air quality models require extensive computational capability, their application is constrained by lack of knowledge of flow patterns in enclosed environments. While promising, these types of modeling methods are not used for estimating exposure to pollutants, rather models based on mass-balance principles are used and will be addressed below.

Mass-balance based indoor air quality models assume uniformly mixed conditions within an effective volume κV, where κ is a proportionality constant, rather than the total, structural, volume V (see Appendix A). Indoor air quality models use the following variables as inputs for estimating indoor air pollutant concentrations: outdoor pollutant levels, multiple indoor sources and their emission rates and indoor activity. Occupant activity indoors affects indoor source emission rates, building air exchange rate, and effective volume, as well as flow rates of cleaning device(s), if present, and their efficiency, a function of their maintenance status (Moschandreas, 1995). The heating ventilating and air conditioning (HVAC) system of a building affects indoor air quality both positively and negatively. It serves as a pollution control device by bringing fresh air from outdoors and thus diluting indoor-generated pollutant levels; it also filters and absorbs some contaminants. The HVAC system may degrade indoor air quality if it is not maintained properly and is unable to perform at design levels. Under certain conditions the HVAC system provides nutrients for bio-pollutants and becomes a source of pollution indoors. Several models are capable of estimating air exchange rates of a building, and, others, emission rates of indoor sources within a building (Moschandreas, 1995). A general indoor air model is shown in Appendix A.

Methodological questions specifically associated with developing countries include concerns of exfiltration, semi-enclosed spaces, plume buoyancy and mixing, and natural dust and resuspension and degradation of building materials. It is common to observe smoke leaking from a house in a developing country because of the comparatively high loading of PM within small spaces, the buoyancy of the plume from the cookstove, high air exchange rates (about 10 h^{-1}), and the open architecture of the kitchen (including highly porous building materials) that are found in many developing country homes. A portion of this smoke infiltrates into the same house immediately without mixing to any significant degree with the outdoor air. Another portion of the smoke mixes with the outdoor air and then infiltrates into the same house. These conditions need to be modeled differently from situations found in developed countries. Properly modified multi-compartment models that treat the near ambient air as a compartment, the box within a box approach, may be a practical alternative. It would be desirable to validate these models under both controlled conditions (test chambers) and field conditions, thereby removing the influence of ETS, resuspension, etc.

A related concern is the smoke leaking from one house and infiltrating into neighboring houses. Developing a model that estimates ambient outdoor concentration near the house would be very useful in assessing risk, source apportionment and predicting overall exposure reduction benefits of cookstove related interventions. Such a model would focus on an individual house or on the center of a housing cluster and estimate levels at breathing height due to the cumulative effect of many houses in a cluster and the indoor concentration in the houses. Such a model may predict that population exposure may not reduce significantly unless every house in a cluster switches to cleaner fuels/stoves. Additional complexity arises from the fact that houses in a cluster use different fuel and cook at different times. For example, kerosene-using houses may experience high exposure levels as compared to wood using houses. Such conditions have not yet been modeled in developing countries, though studies have attempted to measure the extent to which ambient air quality within a cluster can deteriorate during cooking periods (Naeher et al., 1999). Finally, an entire cluster of biomass using houses could contribute significantly to urban pollution levels.

Occupants of many rural and urban poor homes in developing countries cook in spaces that are not enclosed by four walls and a ceiling. Kitchens have three walls with or without a roof (i.e., the entrance is entirely open), or the kitchen may have four walls with a roof of plastic, tin or thatch. Frequently, these sheets are placed few inches above the top of the walls with a gap between the walls and the roof. The "indoor" volume and the exceedingly high air exchange rates above 50 h^{-1} of such structures pose special problems for indoor air quality modeling. Models used for conventional houses will not simulate these conditions.

An easily visible and demarcated plume emanates as a vertical column from the cookstove; within the kitchen, it rises a few feet before dispersing horizontally. Only anecdotal evidence is available on these observations. Typically, the vertical region from the floor, the base-level of the cookstove, to about 3 ft is relatively less polluted. A layer of air 2 ft above this lower region has the highest indoor concentration, the region above that has moderate level concentrations. The exposure consequence of this is that in many situations the woman cook who squats on the ground just next to the stove is perhaps exposed to lower levels of smoke than those standing or sitting in the middle region of the room. Recently Saksena (1999) observed that personal RSP concentration ($d_{50} = 5$ μm) of a sample of mothers who were cooking was significantly less than that of the concentration measured simultaneously by stationary samplers placed near their infants about 1 m from the wood stove. This pattern was not observed in the homes of kerosene users. Thus, there is a need to develop models, that can include the plume effect and imperfect mixing.

Natural dust and resuspension and degradation of building materials are important sources because they add complexity and reduce our ability to interpret data collected from combustion related conditions. Results from a study of 80 urban slum households (Saksena, 1999) indicate that indoor background levels of RSP have a mean value of 200 μg/m^3 in houses with no major indoor combustion or other emission sources of RSP. Clearly, these sources must be incorporated in indoor modeling efforts.

2.1.6. Occupational and in-transit exposures

The type of indoor model discussed above can be used in non-industrial work environments such as offices. The investigator must be cognizant of the size of the building and assure that the volumes assumed to constitute a uniform microenvironment are not too large, are served by the same air-handling unit and have no unusual sources of emission. If the model configuration is more complex one may wish to employ models specifically constructed for office buildings.

Industrial indoor environments are subject to more rigorous regulation than non-industrial work environments. Additionally, industrial environments are dominated by one or few sources that are well defined; measurements are made when it is considered appropriate. Information on levels in industrial environments may be found in the literature and can be obtained from the Occupational Safety and Health Administration (OSHA) and can be used in estimating exposures. When indoor sources emit, estimated or measured emission rates are used as input variables to indoor models. There is an in-transit component of exposure to pollutants, but indoor air models specifically for cars, trucks and buses are not available. While the volume of the in-transit microenvironment is small, the ventilation rate as the sum of mechanical, natural and infiltration is large for automobiles and trains. A first order approximation may be the use of outdoor concentrations as a surrogate for in-transit levels.

2.2. Receptor model estimates of concentrations and source contributions

Microenvironmental concentrations can be measured with area sampling. When chemical components of PM are measured that come from different source types, receptor models can be used to estimate source contributions, see article by Watson and coworkers in this issue.

True receptor models are not statistical black boxes. They are based on the same scientific principles as source models, but they are explanatory rather than predictive of source contributions. Receptor models include the enrichment factors (EF), chemical mass balance (CMB), eigenvector analysis (also termed principal component analysis (PCA), factor analysis (FA), and empirical orthogonal functions), multiple linear regression (MLR), neural networks, cluster analysis, Fourier Transform time series, and a number of other multivariate data analysis methods. Chemical models embedded in source-oriented models can also be used as receptor models to estimate how emissions characteristics might change between source and receptor and to determine limiting precursors for secondary aerosols that form during transport.

2.2.1. Source profiles

Receptor models do not require the spatial and temporal resolution for emissions rates that are needed for source models. They do require an identification of outdoor and indoor source types and the chemical or physical properties that are believed to distinguish their contributions once they arrive at a receptor. The emissions rates described above must be supplemented with these source profiles to apply receptor models. Source profiles are the mass abundances (fraction of total mass) of a chemical species in source emissions. Source profiles are intended to represent a category of source rather than individual emitters. The number and meaning of these categories is limited by the degree of similarity between the profiles.

Chemical or physical properties that are believed to distinguish among different source types are measured on representative emitters. A large variety of chemical and physical components can be measured in source and receptor samples. The sampling methods, flow rates, sample duration, and substrates for microenvironmental and source sampling must be selected (Chow, 1995). Elements (Watson et al., 1999), ions (Chow and Watson,

1999; Chow et al., 2000), and carbon fractions (Chow et al., 1993) are commonly measured.

In geological material, aluminum, silicon, potassium, calcium, and iron have large abundances with low variabilities. The total potassium abundance is 15–30 times the abundance of soluble potassium. Lead is sometimes abundant in paved road dust, probably due to deposition from previously emitted leaded-gasoline vehicle exhaust. Elemental carbon (EC) abundances are highly variable in geological material, and are often negligible in natural soil samples. Organic carbon (OC) is typically 5–15% in geological emitters. Motor vehicle emissions (e.g., brake and tire wear, oil drips) could result in greater abundances of lead, EC, and OC in paved road dust. Soluble sulfate, nitrate, and ammonium abundances are low, in the range of 0–0.3%. Sodium and chloride are also low, with less than 0.5% in abundance. Larger abundances of these materials may be found temporarily soon after roadway deicing, however.

In vehicle exhaust, OC and EC are the most abundant species in motor vehicle exhaust, accounting for over 95% of the total mass. The abundances of organic and total carbon can be quite variable in motor vehicle exhaust profiles. Lead, bromine, and chloride are good markers for gasoline exhaust in areas where leaded fuels are used, but lead is quickly being eliminated in most areas and is completely gone from fuels used in the United States.

Organic and elemental carbons are also abundant in burning and cooking, but the OC fraction is much larger than EC when compared to the vehicle exhaust profiles. The soluble to total potassium ratios of 0.80–0.90 in burning profiles (Calloway et al., 1989) are in large contrast to the low soluble to total potassium ratios found in geological material.

Coal-fired power generation profiles differ substantially from residential coal burning, even though the fuels are similar, owing to the different emission control technologies. Sulfate is one of the most abundant constituents in the particle phase and sulfur dioxide levels can be hundreds to thousands of time higher than the particle mass. Sulfur dioxide is a good indicator of contributions from nearby coal-fired power stations for which it has not reacted or deposited significantly during transport to a receptor. Crustal elements such as silicon, calcium, and iron in the coal-fired boiler profiles are present at 30–50% of the corresponding levels in geological material with the exception of aluminum (Al) which is present at similar or higher levels than those found in geological material. Other elements such as phosphorus, potassium, titanium, chromium, manganese, strontium, zirconium, and barium are present at less than 1% levels. Selenium is often a good indicator of coal-fired power station emissions with no scrubbers or wet scrubbers, but not in emissions from a unit with a dry limestone scrubber. Selenium is usually in the gaseous phase within hot stack emissions, and it condenses on particles when air is cooled in the dilution chamber.

There are similarities in chemical compositions for different sources. However, using source profiles from one airshed or time period may not provide a valid receptor model apportionment for microenvironmental samples in another area or in another time period. Source emissions of precursor gaseous and primary particles are highly variable due to differences in fuel use, operating conditions, and sampling methods. Source and ambient measurements must be paired in time to establish reasonable estimates of source/receptor relationships.

Elemental measurements by themselves are necessary, but insufficient, for a receptor modeling study. Chemical speciation must also include ammonium, sulfate, nitrate, OC, and EC. Simultaneous gas measurements as well as other characteristics of suspended particles such as organic compounds, isotopic abundances, and single particle characteristics are to better-define source categories.

2.2.2. Receptor measurements

Outdoor and indoor samples are often taken only to determine the mass of particle loadings, and these samples are not always amenable to the chemical analyses needed for receptor modeling. A material balance is a preliminary source apportionment that allocates TSP, PM_{10}, or $PM_{2.5}$ to geological material, OC, EC, sulfate, nitrate, ammonium, and possibly salt (from marine aerosol, deicing, or windblown dry lake beds). Although not a source apportionment, the material balance provides guidance on which components are the major cause of excessive mass concentration. This knowledge can focus efforts to improve the emissions inventory for source modeling.

Quartz fiber filter samples that are commonly taken in outdoor and indoor studies are sub-optimal for chemical analysis, but they are commonly available throughout the world. They can be analyzed for elements, ions and carbon, although their background levels may result in degraded detection limits and the inability to detect some important elements such as aluminum and silicon. Volatile components such as ammonium nitrate often evaporate from these samples if they are not adequately maintained. If it is known that chemical analyses will be applied to some or all samples, filters can be acceptance tested prior to use and stored under refrigeration after sampling to minimize interferences and maintain sample integrity. Additional samplers with filter media appropriate for other analyses can be co-located with the compliance sampler. Mini-volume ambient samplers or personal exposure samplers have been adapted for this use.

2.2.3. Receptor model application

All of the multi-variate models include, implicitly or explicitly, a CMB that quantifies source contributions. The CMB (Watson et al., 1984, 1990, 1991) expresses each aerosol property concentration measured at a receptor as the sum of a source contribution multiplied by the abundance of the corresponding aerosol property in the source emissions. When these equations are not collinear (i.e., source profiles substantially differ among different emitters) and the chemical abundances in the source profile are reasonably constant (i.e., standard deviations less than half the average abundance for representative source tests), these equations can be solved for the source contributions. Source profile abundances and the receptor concentrations, with appropriate uncertainty estimates, serve as input data to the CMB model. The output consists of the amount contributed by each source type represented by a profile of the total mass and of each chemical species.

The CMB modeling procedure requires: (1) identification of the contributing source types; (2) selection of chemical species or other properties to be included in the calculation; (3) estimation of the fraction of each of the chemical species which is contained in each source type (source profiles); (4) estimation of the uncertainty in both ambient concentrations and source profiles; and (5) solution of the CMB equations. The effective variance weighted solution (Watson et al., 1984) is almost universally applied because it: (1) theoretically yields the most likely solutions to the CMB equations, providing model assumptions are met; (2) uses all available chemical measurements, not just so-called "tracer" species; (3) analytically estimates the uncertainty of the source contributions based on precisions of both the ambient concentrations and source profiles; and (4) gives greater influence to chemical species with higher precisions in both the source and receptor measurements than to species with lower precisions.

CMB model assumptions are: (1) compositions of source emissions are constant over the period of ambient and source sampling; (2) chemical species do not react with each other (i.e., they add linearly); (3) all sources with a potential for contributing to the receptor have been identified and have had their emissions characterized; (4) the number of sources or source categories is less than or equal to the number of species; (5) the source profiles are linearly independent of each other; and (6) measurement uncertainties are random, uncorrelated, and normally distributed.

The degree to which these assumptions are met in applications depends to a large extent on the particle and gas properties measured at source and receptor. CMB model performance is examined generically by applying analytical and randomized testing methods, and specifically for each application by following an applications and validation protocol. The six assumptions are fairly restrictive and they will never be totally complied with in actual practice. Fortunately, the CMB model can tolerate reasonable deviations from these assumptions, though these deviations increase the stated uncertainties of the source contribution estimates.

An applications and validation protocol (Watson et al., 1991): (1) determines model applicability; (2) selects a variety of profiles to represent identified contributors; (3) evaluates model outputs and performance measures; (4) identifies and evaluates deviations from model assumptions; (5) identifies and corrects model input deficiencies; (6) verifies consistency and stability of source contribution estimates; and (7) evaluates CMB results with respect to other data analysis and source assessment methods.

Since no model, source or receptor, is a perfect picture of reality the results must be independently challenged. Receptor model source contributions should be consistent between locations and sampling times. Discrepancies between source contributions estimated by source and receptor models must be reconciled. Fig. 3 illustrates a comparison between relative $PM_{2.5}$ source contributions estimated from an emissions inventory and a CMB source apportionment at an urban site in Denver, CO during winter, 1996 (Watson et al., 1998). The emissions inventory gives the impression that fugitive dust is the major contributor, while its relative contribution is much lower in the ambient samples. Secondary ammonium sulfate and ammonium nitrate are significant contributors that are not part of a primary particle inventory. These are only estimated by a chemical transport model that also includes emission rates for sulfur dioxide, oxides of nitrogen, and ammonia. Cold starts are identified by the receptor model but not in the emission inventory, indicating that this source should be included in future inventories. The inventory identifies natural gas combustion and other industrial sources that are not distinguished by the receptor model because they do not have chemical profiles that differ from the other, more dominant contributors.

2.3. Human activity patterns

Tools for collecting information about human activities include surveys, questionnaires, time/activity diaries, automated systems such as global position sensing monitors, and observations. Questionnaires are used to collect human activity data and help determine who is exposed, and the magnitude, frequently, location and source(s) of exposure. Activity data can provide distributions, measures of central tendency and measures of variability of behavior within a population or of an individual over time and space.

Certain information from these instruments can be used as surrogates for exposure measurements. The

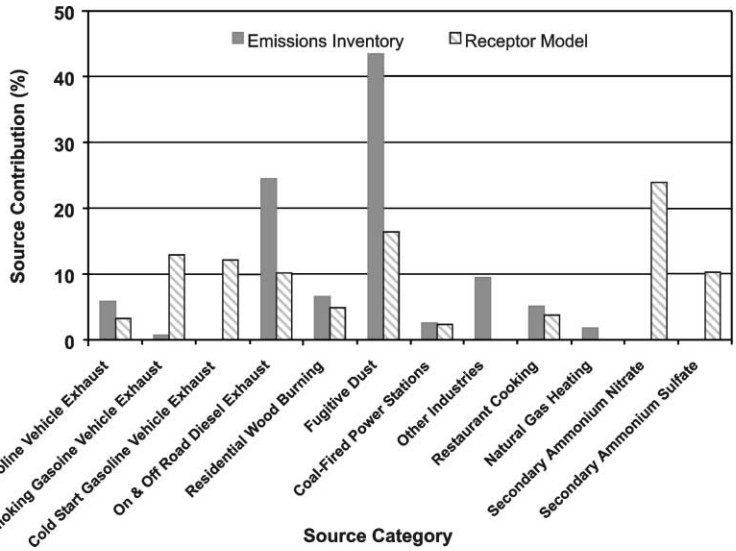

Fig. 3. Comparison of emission inventory and receptor model source apportionment for Wintertime Denver, CO.

number of cigarettes smoked, time spent cooking over an unvented stove, occupation, location and type of heat source in a home, or the distance of a person from diesel exhaust at a bus depot are examples of potential surrogates for exposure. Presence, maintenance status and type of the HVAC system, time spent by an individual in a location or microenvironment and the time of day or season of the year spent in that location are examples of information gathered by questionnaires that relates to factors potentially affecting exposure. If the response to a question(s) is numerical, it is possible to estimate the relative magnitude of an effect on exposure. Categorical groupings are also used in exposure estimates, but categories are often simplistic (yes or no), and subtle degrees of influence of the categorized variable on exposure cannot be determined.

Selection of a specific instrument depends upon the study objective, community characteristics, and budgetary constraints. Brief surveys are useful when large population based studies are conducted and the objective is to characterize the distribution of general exposure variables within the population. Questionnaires and surveys are administered either in person, over the telephone, or by mail when the focus of the desired information is the household and not the individual. Questionnaires and surveys differ only in the level of detail of questions and therefore in the length required for response. Questionnaires provide greater detailed information about exposure activities at one point in time or general information about historical exposures of individuals.

Surveys and questionnaires typically attempt to characterize sources of exposure in terms of occupation, household activities, personal habits, travel activities, and household characteristics. These instruments usually also collect demographic information such as age, gender, race or ethnicity, and some indicator of socioeconomic status (SES) such as household income or education level. SES is often found to be associated with different personal habits such as smoking (Malstrom et al., 1999), with type of heating and cooking fuel, and with other potential sources of exposure, while age has been associated with differences in activity patterns, exposure to air pollutants and ventilation rate (Spier et al., 1992; Shamoo et al., 1994).

Time/activity diaries typically focus on specific activities and exposure related information of the individual. Diaries collect information about where a person spends time. Time/activity diaries collect specific exposure information such as when and where one cooked dinner or the number of cigarettes smoked in each location at what time and so on. By collecting information repeatedly over time, the diary collects multiple snap shots of an individual and can therefore identify the person specific patterns of exposure, including episodic frequency and duration (Freeman et al., 1999). Time activity diaries have helped in understanding human activity patterns (Robinson, 1977; Chapin, 1984; Juster and Stafford, 1985). Time activity diaries are composed of two parts: a time line and a time budget. The time line identifies a person's use of space during the day, typically in terms of at home, at work, and in-transit, with more detail about specific microenvironments such as at home indoors and at home outdoors. The time budget identifies activities that occur throughout the day starting when the person awakens. Often the time budget detail asked is greater than the interests of the study, but may be useful in guiding the participant's memory about

the day's activities. Diaries have replaced short term recall questions in questionnaires since it has been found that recall is often difficult for participants (Robinson, 1985). Diaries, unless administered by an outside party, require literacy on the part of the participant, a sense of time, and usually an extended period of commitment (typically a week) to the study. The extended time period is necessary to evaluate the variations in activities that might occur from day to day or from season to season. This increased participant burden can lead to lower recruitment rates than surveys or interviews that require a one time, short period commitment. However, the wealth of exposure information that can be obtained through a diary may not be obtainable through any other means (NAS, 1991). Often studies use surveys, questionnaires, and time/activity diaries to understand human exposure patterns and use this information in direct and indirect modeling of exposure, and in characterizing the relative distributions of sources and exposures. The American studies NHEXAS and TEAM are such studies.

Direct observation is generally regarded as the most accurate method for assessing behavior because it does not rely on recall, reportage and competency levels of the participant. However, it is labor intensive and requires well-trained observers and researchers to interpret the information. Observations are nearly always combined with other methods, particularly interviews, which may collect survey, questionnaire of time/diary time information. In Bolivia, for example, direct observations are used to develop questionnaires to examine time spent cooking in the morning, in the evening, and the time when fire is started in the morning and is extinguished in the morning, and when it burns off on its own. In developing countries where literacy and concepts of clock time may be poor, the information obtained with surveys, questionnaires and time/activity diary information is collected using trained interviewers, observers or video cameras operated by trained technicians with knowledge about information needed for the study.

3. The direct method of estimating exposure

The most important attribute of this method is that it measures personal exposure and exposure concentrations in several major microenvironments. An equally important aspect of the direct method is its reliance on questionnaires that provide information to be used for establishing relationships between exposure and source, which are then used for formulating risk management strategies. Employing the proper instrumentation (Section 3.1), investigators use the databases to construct regression models (Section 3.2), or stochastic models (Section 3.3). Receptor model measurements and techniques can also be applied to the samples acquired by personal monitors when these samples are amenable to the needed analyses. The receptor model procedures are identical to those described above.

3.1. Measuring methodology

Exposure measurements are performed using small unobtrusive instruments that subjects attach to themselves. This point of contact technique also employs the same type of personal devices as area monitors to measure exposure concentrations in several major microenvironments. Microenvironmental sampling measures integrated personal exposure or exposure concentrations, which may be later used to estimate microenvironmental exposure. The use of information from questionnaires is as important in this method of measurement as in the indirect method. The need for subject cooperation is greater for the direct method of exposure measurement than for the indirect method because the subject is required to carry the sampling device for a relatively long period of time (EPA, 1997).

Exposure measurement techniques are discussed in depth in another chapter of this issue. Several important methodological points are:

1. Personal exposure measuring devices must be unobtrusive, lightweight, robust and accurate. Personal measuring devices that are heavy, noisy and require frequent intervention increase the burden on the subjects and lead to a low response rate that biases the results of a study.
2. Personal exposure and microenvironmental exposure concentrations used for construction of models and association with health effects must reflect the same time period of measurement.
3. A critical element of exposure measurement is the accuracy and precision of personal and area samplers and the quality assurance, quality control program used to generate databases employed for model formulation.

Personal PM_{10}, $PM_{2.5}$ and RSP samples can be collected by established methods using impactor or cyclone type precipitators, 25.37 mm Teflon (or other type) filters and 2–4 l/min battery operated pumps that run for 12–48 h per sample. Exposures of thousands of individuals have already been measured in field studies by such samplers. Depending on the flow rate and the uninterrupted sampling time, the sampling assemblies weigh from 0.5 up to 3.4 kg. The integrated samples are suitable for gravimetric, and depending on the filter material, elemental and chemical analyses. Real time personal PM mass monitors and multi-channel personal particle counters have also been developed and used in

limited studies in the field. They are still expensive, but capable of producing new data of great potential significance. Details on instrumentation appear in separate chapter.

3.2. Regression models

Linear regression models are used to gain insights about the exposure to a pollutant process, and to relate exposure levels to its determinants. The measured personal exposure is the dependent variable, information obtained from the questionnaires and corresponding microenvironmental agent concentrations are the independent variables. The constructed models are tested for model significance and coefficient significance using conventional statistical methods.

Regression model development, using data from direct exposure measurement, follows the conventional model construction process. The example given here is an illustration of the methodology employed by researchers in the field. Personal exposures to respirable PM were measured and modeled to assess implications for air pollution epidemiology (Spengler et al., 1985). Stepwise multiple regression methods were used to construct four predictive models. By choosing only variables with significant, $\alpha = 0.15$, contribution to the model, the first model with $n = 225$ had only one independent variable: ambient RSP, this model was not significant. The second model added to the ambient RSP levels two indicator variables: smoke exposure and employment status. This second model used a sample size of $n = 225$, and explained 16% of the variability in personal exposures, $R^2 = 0.16$. The third model of the process added to the independent variables of model 2 one more variable: time spent at various microenvironments as indicated in subject daily diaries. The microenvironments included were home, work, public places, in-transit and other locations. Only time at work place and public locations contributed significantly as independent variables.

The fourth and most complete model explained 51% of the variability in personal exposure by adding indoor RSP concentrations to the model. This model is illustrated below:

$$E = 8.5 + 0.3\ (\text{ambient RSP}) + 3.1\ (\text{work time}) + 5.4\ (\text{public time}) + 0.6\ (\text{indoor RSP})$$

The final model does not include smoking exposure as an independent parameter; the authors assumed that the inclusion of the indoor variable contains smoke exposure.

By identifying independent variables that contribute to the estimation of personal exposure, regression models associate the personal exposure with potential sources of exposure. This information may be used to investigate associations between source and health effects and, ultimately, to formulate risk management strategies. Importantly, using exposure concentrations of other major microenvironments as the dependent variable similar regression models can be constructed to determine sources contributing to microenvironmental exposures.

Microenvironmental exposures to PM obtained from direct exposure measurement studies have been compared with each other to determine if measurement of one can be used as surrogate for the others. All comparisons of exposure and exposure related information should be performed using values with matching time scales.

3.3. Stochastic models

Regression exposure models simulate exposures by combining pollutant concentrations measured at several microenvironments with time and activity information and assess differences of exposure among individuals, population sub-groups and populations using inferential statistics. Probabilistic or stochastic models also determine inter-individual variability of the predicted parameter and uncertainty about specific statistics of the population distribution. Stochastic models generate a population distribution of exposure to agent(s) and afford the opportunity to compare the population with specific measurement of groups of interest and concern. Knowing the exposure distribution of a population under examination helps respond to the fundamental concern of the relative position of an estimated exposure of specific sub-population against that of the general population. Exposure distributions of sub-populations denoting group characteristics based on geography, ethnic background, age, health status or socioeconomic strata may be compared with the general population exposure distribution and/or specific points of the distribution, (Sexton et al., 1995). Distribution points of importance for comparison purposes include exposures at or about the middle of the distribution or at or above the 90th percentile and the most exposed person.

Sources of exposure uncertainty fall in one or more of three categories, (US EPA, 1992):

1. Scenario uncertainty reflects lack of knowledge to fully specify the problem due to missing or partial information.
2. Parameter uncertainty denotes lack of knowledge regarding the distribution of a model parameter.
3. Model uncertainty is lack of knowledge to fully formulate conceptual and computational models.

Of the three categories of uncertainty, the inhalation exposure model is widely accepted and is not considered a source of uncertainty. The recent development of a new model for estimating exposure to PM, (see the

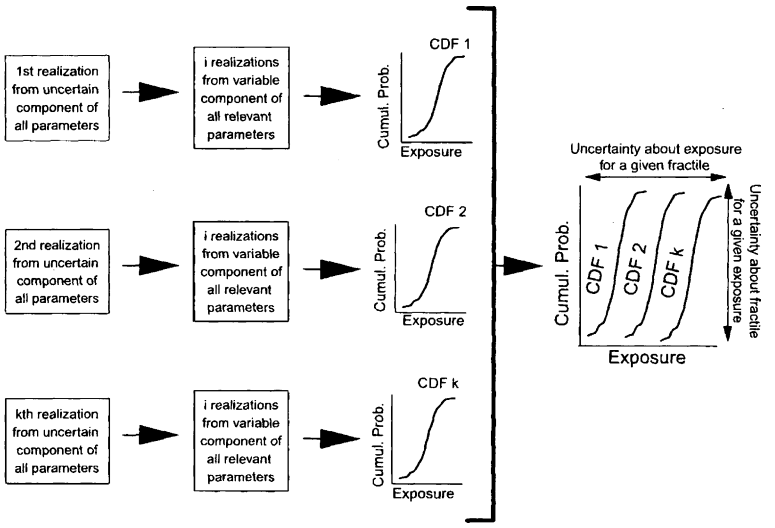

Fig. 4. Illustration of the Monte Carlo simulation method for estimating parameter uncertainty and variability. Source: McIntosh et al. (1995).

introduction section of this chapter) will provide researchers with the opportunity to estimate model uncertainty. Scenario uncertainty can be discussed only when a specific scenario is assessed, thus parameter uncertainty is the focus of discussion in this section.

Parameter uncertainty may be thought of as a composite of two elements: variability and knowledge uncertainty. Variable parameters or *variability* reflect heterogeneity across people, places and time and affects precision of exposure estimates and the degree of their generalization. Knowledge uncertainty, or *uncertainty* reflects lack of knowledge or information about parameters that should be invariant if the perfect measuring means existed. Parameter uncertainty is thought of as the cause of exposure uncertainty and should be investigated in each exposure modeling effort.

Probabilistic uncertainty analysis with Monte Carlo simulations assigns probability density functions to each parameter and then selects values from these distributions into the exposure model under consideration. For inhalation exposure to PM the distribution of each of the two variables of the classical inhalation exposure equation is formulated. Thus, microenvironmental, and or personal concentration and time budget distributions formulated from direct exposure measurement databases are input distributions to a stochastic model for estimating exposure to PM.

The initiating step in stochastic model formulation is to use input to the model variable measurements and construct either "custom" distributions or curve-fit these measurements with best-fitted theoretical distribution function. The procedure for formulating a probabilistic model is a two-phase process (Bogen and Spear, 1987; McIntosh et al., 1995). A simulation is carried out by choosing one realization from the distribution of each uncertain parameter and then, in the second phase, selecting values or realizations repeatedly from all variable parameters. Exposure uncertainty is estimated for a percentile or other statistic from a family of several simulations, each of which is composed of many iterations. This method is illustrated in Fig. 4 (McIntosh et al., 1995). The process generates 500 simulations, each having 1000 iterations, to predict uncertainty about any percentile of the population distribution. A series of efforts are underway to use probabilistic models for assessing uncertainty in inhalation exposure to PM estimations, but none has been published as of this time.

4. Discussion and conclusions

Data for an exposure assessment are obtained from several sources. The indirect method of estimating exposure uses source information from public records, regulatory agencies, and literature, and obtains meteorological data from existing relevant and representative databases or generates meteorological information using models. Exposure studies collect information on subjects using questionnaires. Such information must be representative of the domain of inquiry and is subject to the scale of available information; experimental design considerations focus on obtaining representative sample information of population demographic, time budget, and activity characteristics using questionnaires.

Estimated PM concentrations in microenvironments are combined with time spent by individuals at each corresponding microenvironment to estimate exposure to PM (see Fig. 2). Computationally, this step of the

process is easy, yet the interpretation of the exposure levels generated is not as straightforward. A critical element of exposure estimation is selection of a representative sample of the subject population. Experimental design issues are applicable to exposure estimations or measurements for all exposure methodologies.

The experimental design is formulated to satisfy clearly stated study objectives including criteria of accuracy and precision of estimates, the measuring instrumentation, stationary and personal samplers, questionnaires, models and input to the models variables. A carefully selected experimental design is employed to overcome difficulties associated with measuring and estimating exposures of a subject population. Samples of convenience using volunteers, friends or neighbors restrict the conclusions of a study. Such studies help test specific hypotheses but do not provide a basis for formulating distributions of exposures and reaching conclusions on the population. Probability based samples or representative samples provide the desired unbiased information efficiently.

The objectives of each study determine the exact experimental design; yet the complexity, heterogeneity and variability of exposure to agents within a population, (individuals, time and space) are attributes shared by almost all exposure experimental designs. Such attributes must be characterized using a representative sample and an appropriate sample size. Design issues associated with formulating a national exposure assessment study are elaborated by Callahan et al. (1995). They discuss factors to be considered in designing a national exposure study to multiple pollutants and multiple media determination of sample size as a function of sample design effect and other factors is discussed in detail. The conclusion reached is that a multi-stage probability sampling procedure appears to be applicable for studies of exposure to multiple pollutant, such as the NHEXAS study, or for studies of a class of pollutants, such as the PTEAM study. Further discussion of experimental design is beyond the scope of this paper; yet the implications of each exposure assessment clearly depend on the experimental design, which must coordinate and match the requirements of environmental health studies that are usually associated.

Exposure models have been developed using either direct exposure measurements or indirect exposure estimations. These models assist investigators in estimating exposure to PM and its constituents as a function of a number of independent variables. Furthermore, these models assist in listing and ranking the determinants of exposure to PM and in developing risk reduction strategies.

Exposure assessment advocates assert that the pertinent variable that links source with health effects is the exposure. Exposure studies, therefore, should be designed to assist epidemiology investigations. A critical element of this association is the use of corresponding averaging times, for short term effects require short term sampling, while long term effects should be based on long term averaging of exposures. Current scientific inquiry attempts to gain insights on whether ambient PM concentrations, measured for regulatory purposes, constitute a surrogate for exposure to PM. Conventional exposure measurements and exposure models may be and are used to gain insights into to this inquiry. The alternative approach of categorizing exposure to PM by its general source category is advocated as the exposure methodology that may establish the association between exposure to PM and health effects which has not yet been established by the other methods of exposure analyses.

Source apportionment of personal exposure to PM is a potentially powerful policy tool that combines exposure estimates or measures with information from time activity data and source information derived from questionnaires to construct regression models and other tools in order to relate exposure to PM with its sources. Combining exposure assessment and receptor source modeling has been called total exposure and significant source assessment (TESSA) (Smith and Eglerton, 1990). Basically, this involves receptor–source modeling using personal or time-weighted microenvironmental filters to perform exposure apportionment. Such an approach allows determination of the relative contributions of each of the significant sources categories to the daily exposure of representative members of the population, rather than just the relative contribution of different source categories to concentration at a particular static receptor, as in conventional receptor–source modeling.

Exposure can be reduced by reducing excessive concentrations or by changing the times and locations where people are exposed. The latter option is seldom available except for a few very hazardous industrial occupations. Reducing exposure concentrations requires reduction of indoor and outdoor source emissions. Source contributions to exposure concentrations are estimated by both source and receptor models. Both models should be applied, where possible, as they provide independent concentration estimates that can be used to validate each other. Discrepancies between the two independent source contribution estimates often help to indicate the nature of the discrepancy. Previous studies have found that most discrepancies are caused by inaccuracies in emission inventories against which the amounts of required reductions are measured. A reconciliation of these discrepancies leads to a greater reduction in exposure concentrations per cost unit of a pollution control measure.

Much of the cost of an exposure study depends on the degree to which estimates are aggregated in space, time, and chemical specificity. Measurements at a relatively small number of outdoor sites and indoor micro-

environments must be extrapolated to a larger number of location types. Particle sampling on filters are typically acquired at non-sequential intervals (e.g., every sixth day) and for durations of 24 h or longer. Particle mass concentration is less costly to measure than detailed chemical composition, even though it is believed that certain chemicals have more deleterious effects than others. Although source modeling achieves better resolution, it suffers from inaccuracies in the absence of some measurements over which to evaluate its results. Exposure studies that use available data or reasonable enhancements to those data typically have substantial aggregation with respect to populations, exposure time, the number of microenvironments represented, and the chemical constituents of the particles. These results may be sufficient to make good decisions, but the degree of aggregation must be recognized and described relative to the decisions being made.

Empirical exposure models have been formulated using databases from exposure studies developed under certain environmental, cultural and ethnic conditions. Care must be exercised when these models are used for conditions different from the ones used to formulate the models. It is recommended that properly designed studies be performed to estimate the magnitude of relevant variables under different environmental, cultural and ethnic conditions. Examples of such needs include the determination of building air exchange rates, of emission rates of indoor sources such as cooking facilities, of time budget and time activities in individual countries and others.

Exposure estimates can be made with existing data and with minimal enhancements to those data. Most areas have at least some estimate of emissions rates, some measurements of ambient concentrations, and some knowledge of transport meteorology that can be used to estimate receptor concentrations and the source emissions that cause them. Prognostic meteorological models can supplement existing wind, temperature and humidity measurements to estimate transport and mixing. Particle filters acquired from monitoring networks can be analyzed for some elements, ions, and carbon to obtain a material balance that indicates and verifies emissions inventories. These data can be combined into a conceptual model that can *guide* immediate decision-making and determine the extent to which additional data must be acquired.

Independent observational data or methods should be used to evaluate the performance of exposure assessments as a standard element of the study. For example, source modeling predictions of source category contributions can be checked with application of receptor modeling techniques using independent observational data; indirect exposure assessment predictions can be compared to available direct exposure data; assumptions regarding the region of representativeness of direct data can be evaluated against indirect model predictions.

A reconciliation analysis should be performed to identify and address weakness of previous modeling studies or iterations. The discrepancies between model predictions and observations yield valuable information regarding shortcomings of the analysis. Iterative attempts should be made to improve model performance by applying the information learned in previous studies.

Although the definition of an approach that is well-balanced and consistent with the program resources is necessary, it is essential that the methodology meets a minimum level of performance. It is necessary to understand and identify the main issues needing to be addressed, and then design a modeling approach to meet those needs. The use of a modeling approach designed solely to fit available study resources, without consideration of the technical relevance of the techniques is unacceptable.

Measurement is the preferred means to obtain data. However, it is often the case in developing countries that these measurements do not exist or are not representative or of adequate quality. Often new measurement programs are too expensive and time-consuming to conduct. It is feasible to mathematically simulate some of the data requirements for use in modeling. These simulations can include generation of an emissions database based on mass-balance data or a meteorological database derived from NWP model outputs. Data from these sources are relatively inexpensive, fast to generate and have a more comprehensive coverage (spatial and temporal) than measured data.

Every element of an exposure estimate needs to be balanced with other elements of the study in terms its level of detail, complexity and associated uncertainty. For example, a very detailed treatment in an individual module within a modeling study may not improve the overall uncertainty of the study when large uncertainties exist in other elements of the study. Sensitivity analyses can be used to estimate the relative level of uncertainty associated with individual elements of the study, and resources focused on those elements where the greatest benefit obtained.

Technological advances in the field should be utilized to enable the practitioner to use the most up-to-date tools and methodology to determine exposure assessments. For example, models with limited applicability should not be utilized if there are more sophisticated, accurate models available with which to conduct the study. This will save valuable time and resources and will furthermore minimize any errors or interpretation problems that arise when technologically dated models are used to determine exposure concentrations.

Exposure models were formulated from data obtained from either the indirect or from the direct method of estimating exposure to PM and its constituents. Both

of these methods require information obtained from questionnaire, the difference relates to the methodology of obtaining exposure data. The indirect method is less expensive, but it is limited by the very fact that it generates data from very limited information and few measurements. The direct method uses measurements, but the generation of data is expensive and requires subject participation, which is a source of concern regarding the experimental design and analysis. To optimize cost and accuracy, it is recommended that investigators employ both methods simultaneously, when possible, and use measurements from the direct method to verify estimations of the indirect method.

Appendix A. Model formulations

Following are conceptual formulations for exposure, source, receptor, and emissions models. Models inputs and model outputs are identified, as are the indices for pollutant, source type, time period, subject, receptor location, and individual source type. These indices are indicated for all models as follows:

i	pollutant
l	subject
j	source type
m	receptor location
k	time period
n	source sub-type

Exposure model

$$E_{il} = \sum_{k=1}^{K\text{outdoor}} C_{ikm}t_{klm} + \sum_{k=1}^{K\text{home}} C_{ikm}t_{klm} + \sum_{k=1}^{K\text{transportation}} C_{ikm}t_{klm} + \sum_{k=1}^{K\text{workplace}} C_{ikm}t_{klm}$$

Output

E_{il}	exposure of subject l to pollutant i

Input

C_{ik}	concentration of pollutant i over time period k at location m for subject l
t_{klm}	time period k spent by subject l at location m

Source model

$$C_{ikm} = \sum_{j=1}^{J}\sum_{n=1}^{N} D_{jkn}T_{ijkn}F_{ij}Q_{jn}$$

Output

C_{ikm}	concentration of chemical species i for time period k at location l

Input

D_{jkn}	dispersion and mixing of emissions between source jn and location m over for time period k
T_{ijkn}	transformation of pollutant i between source jn and location m corresponding to time period k
F_{ij}	fractional quantity of pollutant i in source type j
Q_{jn}	total emissions from source jn

Receptor model

$$C_{ikm} = \sum_{j=1}^{J} F_{ij}S_{jkm}$$

$$S_{jk} = \sum_{n=1}^{N} D_{jkn}T_{ijkn}Q_{jn}$$

Output

S_{jkm}	contribution from source type j at location m for time period k
C_{ikm}	concentration of chemical species i for time period k at location l
F_{ij}	fractional quantity of pollutant i in source type j

Emissions model

$$E_{jkl} = R_{jkn}K_{jkn}A_{jkn}(1 - P_{jkn})$$

Output

Q_{jkn}	emissions rate from source type j corresponding to time period k and area n

Input

R_{jkn}	rate of emissions (emissions factor) for a specific size fraction per unity of activity for source type j corresponding to time period k and location of sub-type n
K_{jkn}	particle size reduction applied to R_{jkn} when E_{jkn} is intended to represent a particle size fraction different from that represented by R_{jkn} (e.g., when $PM_{2.5}$ emissions are desired and emissions factors are only available for PM_{10} or TSP). This factor is likely to be different for different source types j, exposure periods k, and locations of sub-type n
A_{jkn}	activity that causes dust emissions for source type j over corresponding to time period k for source sub-type n
P_{jkn}	fractional reduction due to emissions controls applied to source j over time period k and location of sub-type n

Each of the components of Q_{jkn} is empirically derived from a limited number of tests. These tests are intended to represent the entire population of emission factors, activity levels, size distributions, and emissions reduc-

tion effectiveness. Averaging periods are typically for a year or season and averaging areas are typically the sizes of counties or states. Each of these components of fugitive dust emission rate contains uncertainties when applied to a specific situation.

References

Bare, J.C., 1988. Indoor air pollution source database. Journal of the Air Pollution Control Association 38, 670–671.

Barth, D., 1970. Federal motor vehicle emissions goals for CO, HC, and NO_x based on desired air quality levels. Journal of the Air Pollution Control Association 20, 519–527.

Bogen, K.T., Spear, R.C., 1987. Integrating uncertainty and inter-individual variability in environmental risk assessment. Risk Analysis 7 (4), 427–436.

Callahan, M.A., Clinker, R.P., Whitmore, R.W., Kalton, G., Sexton, K., 1995. Overview of important design issues for a national exposure assessment survey. Journal of Exposure Analysis and Environmental Epidemiology 5 (3), 257–282.

Calloway, C.P., Li, S.M., Buchanan, J.W., Stevens, R.K., 1989. A refinement of the potassium tracer method for residential wood smoke. Atmospheric Environment 23, 67–69.

Cass, G.R., 1981. Sulfate air quality control strategy design. Atmospheric Environment 15, 1227–1249.

Cass, G.R., McRae, G.J., 1981. Minimizing the cost of air pollution control. Environmental Science and Technology 15, 748–757.

Cass, G.R., McRae, G.J., 1983. Source–receptor reconciliation of routine air monitoring data for trace metals: An emission inventory assisted approach. Environmental Science and Technology 17, 129–139.

Chapin, L., 1984. Human Activity Patterns. John Wiley Press, New York, NY.

Chow, J.C., 1995. Critical review: Measurement methods to determine compliance with ambient air quality standards for suspended particles. Journal of Air and Waste Management Association 45, 320–382.

Chow, J.C., Watson, J.G., 1999. Ion chromatography. In: Landsberger, S., Creatchman, M. (Eds.), Elemental Analysis of Airborne Particles. Gordon and Breach, Newark, NJ, pp. 97–137.

Chow, J.C., Watson, J.G., Hackett, E.I., Stone, R.H., Hinsvark, B.A., 2000. General atomic absorption procedure for trace metals in airborne material collected on filters. In: Lodge, J.P. (Ed.), Methods of Air Sampling and Analysis, fourth ed. Air and Waste Management Association, Pittsburgh, PA.

Chow, J.C., Watson, J.G., Pritchett, L.C., Pierson, W.R., Frazier, C.A., Purcell, R.G., 1993. The DRI thermal/optical reflectance carbon analysis system: description, evaluation and applications in US air quality studies. Atmospheric Environment 27A, 1185–1201.

deNevers, N., Morris, J.R., 1975. Rollback modeling: Basic and modified. Journal of the Air Pollution Control Association 25, 943–947.

Freeman, N.C.G., Lioy, P.J., Pellizzari, E., Zelon, H., Thomas, K., Clayton, A., Quackenboss, J., 1999. Responses to the Region 5 NHEXAS time/activity diary. Journal of Exposure Analysis and Environmental Epidemiology 9, 414–426.

Grell, G.A., Dudhia, J., Stauffer, D.R., 1994. A description of the fifth generation penn State/NCAR mesoscale model (MM5). NCAR Technical Note NCAR/TN-398+STR, National Center for Atmospheric Research, Boulder, CO. 122pp.

Juster, F.T., Stafford, F.P., 1985. Time, Goods, and Well-being. University of Michigan Survey Research Center, Ann Arbor, MI.

Kim, N.D., Fergusson, J.E., 1993. Concentrations and sources of cadmium, copper, lead and zinc in house dust in Christchurch, New Zealand. Science of the Total Environment 138, 1–22.

Koutrakis, P., Spengler, J.D., Chang, B., Ozkaynak, H., 1987. Characterizing sources of indoor and outdoor aerosols using PIXE. Nuclear Instruments and Methods in Physics Research B 22, 331–336.

Lewis, C.W., 1989. Sources of air pollutants indoors. Atmospheric Environment, 312–322.

Lewis, C.W., 1991. Sources of air pollutants indoors: VOC and fine particulate species. Journal of Exposure Analysis and Environmental Epidemiology 1, 31–44.

Lionel, T., Martin, R.J., Brown, N.J., 1986. A comparative study of combustion in kerosene heaters. Environmental Science and Technology 20, 78–85.

Lioy, P.J., Wainman, T., Zhang, J., Goldsmith, S., 1999. Typical household vacuum cleaners: The collection efficiency and emissions characteristics for fine particles. Journal of Air and Waste Management Association 49, 200–206.

Lofroth, G., Stensman, C., Brandhorst-Satzkorn, M., 1991. Indoor sources of mutagenic aerosol particulate matter: Smoking, cooking and incense burning. Mutation Research 261, 21–28.

Malstrom, M., Sundquist, J., Johansson, S.E., 1999. Neighborhood environment and self-reported health status: A multilevel analysis. American Journal of Public Health 89, 1186–1189.

McIntosh, D.L., Xue, J., Ozkayanak, H., Spengler, J., Ryan, P.B., 1995. A population-based exposure model for benzene. Journal of Exposure Analysis and Environmental Epidemiology 5 (3), 375–403.

Moschandreas, D.J., 1995. Dynamics of Indoor Air Contaminants in Indoor Air Quality. Elsevier, Amsterdam.

Naeher, L.P., Smith, K.R., Leaderer, B.P., 1999. Indoor and outdoor $PM_{2.5}$ and CO transects during cooking in high- and low-density Guatemalan villages. In: Indoor Air' 99. Organizing Committee, (Ed.), Proceedings of the 8th International Conference on Indoor Air Quality and Climate, Edinburgh, UK.

National Academy of Sciences (NAS), 1991. Human Exposure Assessment for Airborne Pollutants: Advances and Opportunities. National Academy Press, Washington, DC.

Petersen, W.B., Lavdas, L.G. 1986. INPUFF 2.0—A multiple source Gaussian puff dispersion algorithm. Users Guide. Research Triangle Park, NC: ASRL, US Environmental Protection Agency.

Raiyani, C.V., Shah, S.H., Desai, N.M., 1993. Characterization and problems of indoor air pollution due to cooking stove smoke. Atmospheric Environment 27A, 1643–1655.

Robinson, J.P., 1977. How Americans Use Their Time. Praeger Publishers, New York, NY.

Robinson, J.P., 1985. The validity and reliability of diaries versus alternative time use measures. In: Juster, F.T., Stafford, F.P. (Eds.), Time, Goods, and Well-Being. University of Michigan Survey Research Center, Ann Arbor, MI.

Saksena, S., 1999. Integrated exposure assessment of airborne pollutants in an urban community using biomass and kerosene cooking fuels. Ph.D. Thesis, Indian Institute of Technology, Center for Environmental Sciences and Engineering. Mumbai, India.

Samet, J.M., Spengler, J.D., 1991. Indoor Air Pollution—A Health Perspective. Johns Hopkins University Press, Baltimore, MD.

Schare, S., Smith, K.R., 1995. Particulate emission rates of simple kerosene lamps. Energy for Sustainable Development 2, 32–35.

Schulze, R.H., Turner, D.B., 1996. Practical Guide to Atmospheric Dispersion Modeling. Trinity Consultants Inc., Dallas, TX.

Scire, J.S., Strimaitis, D.S., Yamartino, R.J., 1999. A user's guide for CALPUFF dispersion model (Version 5). Earth Tech, Inc., Concord, MA.

Sexton, K., Kleffman, D.E., Callahan, M.A., 1995. An introduction to the National Human Exposure Assessment Survey (NHEXAS) and related phase I field studies. Journal of Exposure Analysis and Environmental Epidemiology 5, 229–232.

Shamoo, D.A., Kinn, W.S., Peng, R.C., Solomon, J.C., Webb, R.L., Hackney, J.D., Gong, H., 1994. Time-activity patterns and diurnal variation of respiratory status in a panel of asthmatics: Implications for short-term air pollution effects. Journal of Exposure Analysis and Environmental Epidemiology 4, 133–148.

Smith, K.R., Eglerton, S.A., 1990. Exposure-based air pollution control strategies: Proposed three-way integration. In: Bretthauer, E.W. (Ed.), Total Exposure Assessment Methodology, VIP-16. Air and Waste Management Association, Pittsurgh, pp. 630–641.

Spengler, J.D., Treitman, R.D., Tosteson, T.D., Mage, D.T., Soczek, M.L., 1985. Personal exposures to respirable particulates and implications for air pollution epidemiology. Environmental Science and Technology 19 (8), 700–707.

Spier, C.E., Little, D.E., Trim, S.C., Johnson, T.R., Linn, W.S., Hackney, J., 1992. Activity patterns in elementary and high school students exposed in oxidant pollution. Journal of Exposure Analysis and Environmental Epidemiology 2, 277–293.

Turner, D.B., 1970. Workbook of Atmospheric Dispersion Estimates. US Environmental Protection Agency, Research Triangle Park, NC.

Turner, D.B., 1979. Atmospheric dispersion modeling: A critical review. Journal of the Air Pollution Control Association 29, 502–519.

US EPA, 1992. Guidelines for exposure assessment. Federal Register 57, 22888–22938.

US EPA, 1995. User's guide for the industrial source complex (ISC3) dispersion models, vol. II: Description of model algorithms. Report no. EPA-454/B-95-003b, US EPA, Research Triangle Park, NC.

US EPA, 1997. Exposure factors handbook. US Environmental Protection Agency, Research Triangle Park, NC.

US EPA, 1998. National air pollutant emission trends, procedures document, 1900–1996. Report no. EPA-454/R-98-008, US EPA.

US EPA, 1999. Compilation of air pollutant emission factors—vol. I, Stationary point and area sources. Report no. AP-42, 5th ed., US Environmental Protection Agency, Research Triangle Park, NC.

Wallace, L., 1996. Indoor particles: A review. Journal of Air and Waste Management Association 46, 98–126.

Watson, J.G., Chow, J.C., Pace, T.G., 1991. Chemical mass balance. In: Hopke, P.K. (Ed.), Receptor Modeling for Air Quality Management. Elsevier Press, New York, NY, pp. 83–116.

Watson, J.G., Chow, J.C., Frazier, C.A., 1999. X-ray fluorescence analysis of ambient air samples. In: Landsberger, S., Creatchman, M. (Eds.), Elemental Analysis of Airborne Particles. Gordon and Breach, Newark, NJ, pp. 67–96.

Watson, J.G., Cooper, J.A., Huntzicker, J.J., 1984. The effective variance weighting for least squares calculations applied to the mass balance receptor model. Atmospheric Environment 18, 1347–1355.

Watson, J.G., Robinson, N.F., Chow, J.C., Henry, R.C., Kim, B.M., Pace, T.G., Meyer, E.L., Nguyen, Q., 1990. The USEPA/DRI chemical mass balance receptor model, CMB 7.0. Environmental Software 5, 38–49.

Watson et al., 1998.

Wilson, W.E., Mage, D., 1999. Reinventing personal exposure to particulate matter. In: Third Colloquium on Particulate Matter and Health, Durham, North Carolina.

Yamartino, R.J., Strimaitis, D.G., Scire, J.S., Insley, E.M., Spitzak, M.J., 1996. Final report on the phase 1 development of the kinematic simulation particle (KSP) atmospheric dispersion model. Report no. 1274-3m. Prepared for Institiut fuer Meteorologie Freie Universitaet Berlin and Unweldbundemsamt, Berlin, Germany, EarthTech Inc., Concord, MA.

Yocom, J.E., 1982. Critical review: Indoor–outdoor air quality relationships. Journal of the Air Pollution Control Association, 500.

Yocom, J.E., McCarthy, S.M., 1991. Measuring Indoor Air Quality—A Practical Guide. John Wiley & Sons Ltd., Chichester, West Sussex, England.