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Receptor modeling application framework for particle source apportionment

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Abstract

Receptor models infer contributions from particulate matter (PM) source types using multivariate measurements of particle chemical and physical properties. Receptor models complement source models that estimate concentrations from emissions inventories and transport meteorology. Enrichment factor, chemical mass balance, multiple linear regression, eigenvector, edge detection, neural network, aerosol evolution, and aerosol equilibrium models have all been used to solve particulate air quality problems, and more than 500 citations of their theory and application document these uses. While elements, ions, and carbons were often used to apportion TSP, PM_{10}, and $PM_{2.5}$ among many source types, many of these components have been reduced in source emissions such that more complex measurements of carbon fractions, specific organic compounds, single particle characteristics, and isotopic abundances now need to be measured in source and receptor samples. Compliance monitoring networks are not usually designed to obtain data for the observables, locations, and time periods that allow receptor models to be applied. Measurements from existing networks can be used to form conceptual models that allow the needed monitoring network to be optimized. The framework for using receptor models to solve air quality problems consists of: (1) formulating a conceptual model; (2) identifying potential sources; (3) characterizing source emissions; (4) obtaining and analyzing ambient PM samples for major components and source markers; (5) confirming source types with multivariate receptor models; (6) quantifying source contributions with the chemical mass balance; (7) estimating profile changes and the limiting precursor gases for secondary aerosols; and (8) reconciling receptor modeling results with source models, emissions inventories, and receptor data analyses.
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Keywords: Receptor model; PM_{10}; $PM_{2.5}$; Chemical mass balance; Enrichment factor; Principal component analysis; Factor analysis

Contents

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1. Introduction

Receptor models infer contributions from different source types using multivariate measurements taken at one or more receptor locations. These receptors can be fixed indoor or outdoor monitors or mobile samplers that follow the activities of an individual or group of people. Source models estimate receptor concentrations from source emissions and meteorological measurements. Receptor models use ambient concentrations and the abundances of chemical components in source emissions to quantify source contributions. Source and receptor models are complementary rather than competitive. Each has strengths and weaknesses that compensate for the other. Both types of models can and should be used in an air quality source assessment of outdoor and indoor air.

Chemical and physical analysis methods are often termed receptor models. A chemical ''tracer'' is often sought for certain sources. Microscopic analysis, gas chromatograms, X-ray spectra, carbon-14 (^{14}C) and other isotopic abundances, and many other analytical outputs provide patterns that might allow a source contribution to be identified and quantified. Without the receptor model mathematics and applications framework, however, these analytical methods do not quantify source contributions.

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 (EFs), chemical mass balance (CMB), eigenvector analysis (also termed principal component analysis (PCA), factor analysis (FA), and empirical orthogonal functions (EOF)), multiple linear regression, neural networks, edge detection, cluster analysis, Fourier Transform time series, and a number of other multivariate data analysis methods. Chemical models that are usually 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.

Several books (Hopke, 1985, 1991), specialty conference proceedings (Macias and Hopke, 1981; Hopke and Dattner, 1982; Pace, 1986; Watson, 1989; Watson et al., 1989; Chow et al., 1993c), and review articles (Cooper and Watson, 1980; Gordon, 1980, 1988; Watson, 1984; Javitz et al., 1988a; Watson et al., 2001a; Chow and Watson, 2002; Watson and Chow, 2002b) examine different types and variations of receptor models. Table 1 describes several receptor model types and classifies published references by theory and application. Examples are drawn from many countries throughout the world. Some of the receptor model applications apply to indoor air and personal exposure, but most determine contributions to particles in outdoor air. Neural networks, edge detection, and chemical evolution receptor models are still under development and are of greater interest to researchers than to those attempting to solve air pollution problems. EFs and the CMB are well established and have been widely used to develop pollution control strategies. The multivariate PCA, FA, and EOF models have been used to confirm and sometimes identify unrecognized source types; their use for estimating source profiles for input to the CMB is still under study.

The particle size fractions normally monitored throughout the world are total suspended particulate (TSP, mass of particles with aerodynamic diameters $\lesssim 30$ μm), PM_{10} (aerodynamic diameters <10 μm), and $PM_{2.5}$ (aerodynamic diameters <2.5 μm). $PM_{2.5}$ is becoming more commonly measured because this fraction can penetrate deep into the human lung and more efficiently penetrates between outdoor and indoor environments. $PM_{2.5}$ contains most of the carbon, sulfate, nitrate, and heavy metal components, while the larger particles are dominated by suspended dust, with some contributions from sea salt, pollen and plant detritus.

This article provides a framework for using receptor models to determine those sources requiring emissions reductions to improve air quality in populated areas

Table 1
Summary of receptor model types with references to their theory and use

Explanation	Theory and evaluation	Application
Enrichment factors		
The ratios of atmospheric concentrations of elements to a reference element are compared to the same ratios in geological or marine material. Differences are explained in terms of anthropogenic sources. Heavy metal enrichments are usually attributed to industrial emitters. Sulfur enrichment is attributed to secondary sulfate. Potassium enrichment is attributed to burning and cooking. Local soil and road dust compositions often differ from global crystal compositions	Vinogradov (1959), Turekian and Wedepohl (1961), Taylor (1964), Mason (1966), Gordon et al. (1973), Zoller et al. (1974), Rahn (1976), Lawson and Winchester (1979a), Reimann and de Caritat (2000)	Dams et al. (1971), Hoffman and Duce (1972), Hoffman et al. (1972), Moyers and Duce (1972), Tsunogai et al. (1972), Wilkniss and Bressan (1972), Bogen (1973), Bressan et al. (1973), Zoller et al. (1973, 1974), Heindryckx and Dams (1974), Mroz and Zoller (1975), Struempler (1975), King et al. (1976), Neustadter et al. (1976), Meinert and Winchester (1977), Moyers et al. (1977), Rahn et al. (1977), Buat-Ménard and Arnold (1978), Lawson and Winchester (1978, 1979b), Husain and Samson (1979), Fogg and Rahn (1984), Husain et al. (1984), Pacyna (1986), Lowenthal and Rahn (1987b), Sturges (1989), Carter and Borys (1993), Rashid and Griffiths (1993), Benner et al. (1995), Aunela-Tapola et al. (1998), Gatz et al. (1998), Deboudt et al. (1999), Reimann and de Caritat (2000), Chen et al. (2001), Choi et al. (2001), Paciga et al. (1975, 1976), Winchester et al. (1974, 1979)
Chemical mass balance		
Ambient chemical concentrations are expressed as the sum of products of species abundances and source contributions. These equations are solved for the source contributions when ambient concentrations and source profiles are supplied as input. Several different solution methods have been applied, but the effective variance least squares estimation method is most commonly used because it incorporates precision estimates for all of the input data into the solution and propagates these errors to the model outputs	Britt and Luecke (1973), Friedlander (1973a, 1981), Watson (1979), Belsley et al. (1980), Cooper and Watson (1980), Gordon et al. (1981), Core et al. (1982), deCesar and Cooper (1982), Gerlach et al. (1982), Anderson et al. (1984), Dzubay et al. (1984), Stevens and Pace (1984), Daisey (1985), Gordon and Olmez (1986), Holzman et al. (1986), US EPA (1987), Vong et al. (1988), Larson and Vong (1989), Wang and Hopke (1989), Matamala and Nininger (1990), Kim et al. (1992a), Song and Hopke (1996a,b), Gleser (1997), Hopke and Song (1997), Gordon (1980, 1988), Watson et al. (1981, 1984, 1990b, 1991, 1997b, 1998c), Henry (1982, 1992, 1997a), Currie et al. (1984, 1994), deCesar et al. (1985, 1986), Cheng and Hopke (1986, 1989), Lowenthal et al. (1987, 1988, 1992, 1994), Javitz et al. (1988a,b), Lowenthal and Rahn (1988a,b, 1989), Kim and Henry (1989, 1999)	Hidy and Friedlander (1971), Winchester and Nifong (1971), Miller et al. (1972), Kneip et al. (1973), Friedlander (1973a,b), Gartrell and Friedlander (1975), Hammerle and Pierson (1975), Kowalczyk et al. (1978, 1982), Mizohata and Mamuro (1979), Watson (1979), Alpert and Hopke (1980, 1981), Cooper (1980), Dzubay (1980, 1989), Rheingrover and Gordon (1980, 1988), Heisler et al. (1980), Gatz et al. (1981), Stelson and Seinfeld (1981), Houck et al. (1982, 1992), Johnson and McIntyre (1982), Liljestrand (1982), Stolzenburg et al. (1982), Cass and McRae (1983), Feeley and Liljestrand (1983), Rahn and Lowenthal (1984, 1985), Scheff et al. (1984), Stevens et al. (1990), Chow (1985), Lowenthal and Rahn (1985), Mangelson et al. (1985), Sexton et al. (1985), Chow and Spengler (1986), Lewis et al. (1986, 1988), Rau (1986), Eatough et al. (1987, 1992, 1996, 1997a, 1997b, 2000), Iyer et al. (1987), Batterman et al. (1988), Chow et al. (1988b, 1990a,b, 1991a,b, 1992a,b, 1995, 1996b, 1999, 2000), Cooper et al. (1988, 1989), Dresser and Baird (1988), Dzubay et al. (1988), Gray et al. (1988), Hlavinka and Bullin (1988), Magliano (1988), Vong et al. (1988), Casuccio et al. (1989), Dzubay and Mamane (1989), Eatough (1989), Ge et al. (1989), Miller et al. (1989), Rau and Khalil (1989), Stafford and Liljestrand (1989), Vossler et al. (1989), Egami et al. (1990), Kasahara et al. (1990), Kim and Hopke (1990), Pistikopoulos et al. (1990), van Borm et al. (1990), Conner and Stevens (1991), Glover et al. (1991), Toriyama et al. (1991), White and Macias (1991), Yoshizumi (1991), Hawthorne et al. (1992), Heaton et al. (1992), Kim et al. (1992b), Müller (1992), Skidmore and Chow (1992), Vermette et al. (1992), Zelenka et al. (1992, 1993), Annegarn and Pzybylowicz (1993), Wadden et al. (1993), Bighouse and Houck (1994), Sharma and Patil (1994), Sheffield et al. (1994), Venkataraman and Friedlander (1994), Watson et al. (1994b, 1996, 1997a, 1998b, 1999b), Adachi et al. (1995), Kao and Friedlander (1995), Zhu et al. (1995a,b, 1996, 1998, 1999), Lowenthal et al. (1996, 1997a,b), Miguel et al. (1995), Harrison et al. (1996), Hidy and Venkataraman (1996), Schauer et al. (1996), South Coast Air Quality Management District (1996), Chen et al. (1997, 2001), Fujita et al. (1997, 1998), Malm and Gebhart (1997), Vega et al. (1997), Willis et al. (1997), Adgate et al. (1998), Artaxo et al. (1998a,b, 1990), Biegalski et al. (1998), Coe and Chinkin (1998), Liu et al. (1998), Magliano et al. (1998, 1999), Pinto et al. (1998), R.J. Lee Group (1998), Zhang et al. (1998a), Hosiokangas et al. (1999), Pitchford et al. (1999), Brook et al. (2000), Green and Tombach (2000), Engelbrecht et al. (2000, 2001, 2002) Schauer and Cass (2000), Mazzera et al. (2001), Park et al. (2001a,b), Shu et al. (2001), Temesi et al. (2001), Chow and Watson (2002)

Table 1 (*continued*)

Explanation	Theory and evaluation	Application
Linear regression Mass, sulfate concentrations or light extinction are expressed as a linear sum of unknown regression coefficients times source marker concentrations measured at a receptor. The markers must originate only in the source type being apportioned, which is a stringent assumption rarely met in practice. The regression coefficients represent the inverse of the chemical abundance of the marker species in the source emissions. The product of the regression coefficient and the marker concentration for a specific sample is the tracer solution to the CMB that yields the source contribution	Ito et al. (1986), White (1986, 1989), White and Macias (1987), Milionis and Davies (1994a,b), Lowenthal et al. (1995), Ayers (2001)	Kleinman et al. (1980), Ouimette et al. (1981), Ouimette and Flagan (1982), Kneip et al. (1983), Morandi et al. (1987, 1991), Trijonis et al. (1987), Lowenthal and Rahn (1989), Malm et al. (1990b), Okamoto et al. (1990), Malm and Gebhart (1997), Chan et al. (1999), Green and Tombach (2000)
Temporal and spatial correlation eigenvectors (PCA, FA, EOF) Temporal correlations are calculated from a time series of chemical concentrations at one or more locations. Eigenvectors of this correlation matrix are determined and a subset is rotated to maximize and minimize correlations of each factor with each measured species. The factors are interpreted as source profiles by comparison of factor loadings with source measurements. Several different normalization and rotation schemes have been used, but their physical significance has not been established Spatial correlations are calculated from chemical measurements taken on simultaneous samples at a large number of locations. Eigenvectors of this correlation matrix represent a spatial distribution of source influence over the area, providing that the samplers have been located to represent the gradients in source contributions. As with temporal correlation models, several normalization and rotation schemes have been applied	Armstrong (1967), Blifford and Meeker (1967), Prinz and Stratmann (1968), Crawford (1975), Duewer et al. (1976), Gether and Seip (1979), Hopke (1981, 1988), Richman (1981, 1986), Hopke et al. (1982, 1983, 1995), Karl et al. (1982), Bergametti et al. (1983), Cooper (1983), Henry (1984, 1986, 1987, 1991, 1997b), Hwang et al. (1984), Chow and Spengler (1986), Ito et al. (1986), Derde et al. (1987), Heidam (1987), Lowenthal and Rahn (1987a), Cheng and Lioy (1989), Henry and Kim (1989, 1990), Zeng and Hopke (1989, 1992a), Henry et al. (1991, 1999), Glover and Hopke (1992, 1994), Green et al. (1992a), Kuik et al. (1993a), Paatero and Tapper (1993, 1994), Richman and Vermette (1993), Nitta et al. (1994), Poissant (1994), Brunet and Vautard (1996), Centner et al. (1996), Moro et al. (1997), Paatero (1997, 1998, 2000) Brumelis et al. (2000)	Blifford and Meeker (1967), Prinz and Stratmann (1968), Vavilova et al. (1969), Peterson (1970), Padmanabhamarty (1975), Hopke (1976, 1981, 1982), Hopke et al. (1976, 1980, 1982, 1988, 1993), Gaarenstroom et al. (1977), Barone et al. (1978), Gatz (1978), Hardy and Walton (1978), Gether and Seip (1979), Henry and Hidy (1979, 1981, 1982), Alpert and Hopke (1980, 1981), Boutron and Martin (1980), Throgmorton (1980), Cox and Clark (1981), Rohbock et al. (1981), Chang et al. (1982), Houck et al. (1982), Lioy et al. (1982), Liu et al. (1982, 1995), Overland and Preisendorfer (1982), Rahn et al. (1982), Roscoe et al. (1982, 1984), Bergametti et al. (1983), Cheng et al. (1993a,b), Hoogerbrugge et al. (1983), Severin et al. (1983), Ashbaugh et al. (1984), Malm et al. (1985, 1987, 1990a), Smeyers-Verbeke et al. (1984), Pitchford and Pitchford (1985), Thurston and Spengler (1985a,b), Wolff and Korsog (1985), Wolff et al. (1985a,b), Chow and Spengler (1986), Crawley and Sievering (1986), Daisey et al. (1986), Thomas (1986), Johnson and Malm (1987), Keiding et al. (1987), Kim et al. (1987), Koutrakis and Spengler (1987), Kim and Hopke (1988), Lowenthal and Rahn (1987a), Lowenthal et al. (1992), Pratsinis et al. (1988), Valaoras et al. (1988), Henry and Kim (1989), Koutrakis et al. (1989), Pio et al. (1989), Artaxo et al. (1990), Artaxo et al. (1998a, 1990), Bomboi et al. (1990), Gebhart et al. (1990), Okamoto et al. (1990), Henry et al. (1991), Morandi et al. (1991), Bridgman (1992), Buhr et al. (1992, 1995, 1996), Green et al. (1992a,b, 1993), Rouhani et al. (1992), Sharma and Patil (1992), Sharma and Singh (1992), Zeng and Hopke (1992b), Gao et al. (1993a,b), Kuik et al. (1993a,b), Michaud et al. (1993, 1996), Richman and Vermette (1993), Anttila et al. (1994), Berg et al. (1994), Brown and Lund (1994), Nitta et al. (1994), Xie et al. (1994a,b, 1999a,b), Plumb and Zheng (1996), Sjögren et al. (1996), Swietlicki et al. (1996), Veltkamp et al. (1996), Deininger and Saxena (1997), Gebhart and Malm (1997), Henry (1997b,c,d), Yoo and Kim (1997), Echalar et al. (1998), Norris et al. (1998), Statheropoulos et al. (1998), Polissar et al. (1998, 1999, 2001a,b), Currie et al. (1999), Huang et al. (1999, 2001), Lee et al. (1999), Li and Deng (1999), Paterson et al. (1999), Rocha et al. (1999), Yakovleva et al. (1999), Ames et al. (2000), Chan et al. (2000), Chueinta et al. (2000), Kim and Henry (2000), Kusmierczyk-Michulec and Marks (2000), Ogawa et al. (2000), Pryor and Barthelmie (2000), Ramadan et al. (2000), Tsai et al. (2000), Hien et al. (2001), Jeon et al. (2001), Kavouras et al. (2001), Park et al. (2001a), Poirot et al. (2001), Roberts et al. (2001), Song et al. (2001)

Neural networks

Known inputs and outputs are presented to a neural network that simulates the human though process. The network assigns weights to the inputs that reproduce the outputs. Once these patterns have been established for cases where outputs are known, the weights can be applied to input data to estimate outputs. Neural networks can also be used to provide function relationships and represent a solution to the CMB equations

Reibnegger and Weiss (1991), Wienke and Hopke (1994a,b), Wienke et al. (1994a,b, 1995), Song and Hopke (1996a,b), Gardner and Dorling (1998), Reich et al. (1999)

Zelenka et al. (1992, 1994), Boznar et al. (1993), Gao et al. (1994), Wienke et al. (1994a,b, 1995), Xie et al. (1999b,c), Fan et al. (1995), Cheng et al. (1996), Liu et al. (1996), Rege and Tock (1996), Song and Hopke (1996a,b), Comrie (1997), Potukuchi and Wexler (1997), Mihalakakou et al. (1998), Reifman and Feldman (1998), Gardner and Dorling (1999), Reich et al. (1999), Soja and Soja (1999), Song et al. (1999a,b,c), Kao and Huang (2000), Krijnsen et al. (2000), Perez et al. (2000), Silverman and Dracup (2000), Bhave et al. (2001), Pasini et al. (2001)

Edge detection

Edges are constant ratios among chemical components that are detected in multi-dimensional space. The data set must contain many samples with multivariate measurements and several samples in which sources types are both present and absent. The "edges" detected by models such as UMIX, are translated into source profile abundances that are used in an implicit or explicit CMB

Henry (1997a, 2000, 2002)

Willis (2000), Poirot et al. (2001)

Aerosol evolution

Source profiles containing particle chemical components and gaseous precursors are mathematically "aged" using a chemical reaction scheme. Source profile evolution has been done using Lagrangian source models to simulate the conditions that a profile might encounter en route between source and receptor

Grosjean and Friedlander (1975), Bassett et al. (1981), Friedlander (1981), Lewis and Stevens (1985), Stockwell et al. (1988, 1997), Grosjean and Seinfeld (1989), Wexler and Seinfeld (1991), Pandis et al. (1992), Stockwell (1994), Wexler et al. (1994), Bowman et al. (1995), Fraser et al. (1996), Seinfeld (1997), Kuhn et al. (1998), Seinfeld and Pandis (1998), Zhang et al. (1999), Watson et al. (2002)

Grosjean and Friedlander (1975), Roberts and Friedlander (1975), White et al. (1977), Bassett et al. (1981), Friedlander (1981), Russell et al. (1983), Robinson and Whitbeck (1985), Gordon and Olmez (1986), Iyer et al. (1987), Grosjean and Seinfeld (1989), Latimer et al. (1990), Wexler and Seinfeld (1990, 1991), Pandis et al. (1992), Pankow (1993), Stockwell (1994), Venkataraman and Friedlander (1994), Wexler et al. (1994), McDonald et al. (1996), Kleeman et al. (1997), Lurmann et al. (1997), Gray and Cass (1998), Stockwell et al. (1988, 1990, 2000), Kleeman and Cass (1999), West et al. (1999), Capaldo et al. (2000), Eatough et al. (2000), Chen et al. (2002), Tsuang et al. (2002)

Gas/particle equilibrium

The portions of a semi-volatile species in the gas and particle phase are estimated based on receptor measurements. This shows which precursor is in excess and which needs to be diminished in order to reduce

Hänel (1965), Stelson et al. (1979), Stelson and Seinfeld (1982a,b, 1982c), Saxena et al. (1986), Pankow (1987, 1988, 1992, 1994a, 1994b, 1998), Pilinis et al. (1987), Harrison et al. (1990), Harrison and MacKenzie (1990), Pankow and Bidleman (1992),

Stelson et al. (1979), Stelson and Seinfeld (1982a,b, 1982c), Bassett and Seinfeld (1983, 1984), Harrison and Pio (1983), Saxena and Seigneur (1983), Saxena et al. (1986), Pilinis and Seinfeld (1987), Pilinis and Farber (1991), Barrett et al. (1992), Tanner and Harrison (1992), Wexler and Seinfeld (1992), Kim et al. (1993a,b), Koutrakis and Kelly (1993), Mozurkewich (1993), Koloutsou-Vakakis and Rood (1994), Watson et al. (1994c), Kim and Seinfeld (1995), Meng et al. (1995), Hayami and Carmichael (1997),

Table 1 (*continued*)

Explanation	Theory and evaluation	Application
concentrations in the particle phase. The theory is most highly developed for ammonium nitrate and has been used to determine the extent to which ammonia or oxides of nitrogen reductions are needed to reduce ambient ammonium nitrate levels	Kim et al. (1993a,b), Pankow et al. (1993), Harrison and Msibi (1994), Kim and Seinfeld (1995), Meng et al. (1995, 1997), Matsumoto and Tanaka (1996), Meng and Seinfeld (1996), Jang and Kamens (1998), Ansari and Pandis (1999a,b), Lazaridis (1999), Blanchard et al. (2000), Zhang et al. (2000)	Audiffren et al. (1998), Ansari and Pandis (1999a,b), Diaz et al. (1999), Jacobson (1999), Blanchard et al. (2000), Zhang et al. (2000), Clegg et al. (2001), Moya et al. (2001)

with a variety of industrial, area, and mobile emissions. An eight-step receptor source apportionment procedure is outlined and illustrated with examples from wintertime Denver, CO (Watson et al., 1998b). These examples are complemented by information extracted from several of the references in Table 1.

2. Receptor modeling procedure

Semi-quantitative source apportionment can be applied to existing samples from air quality compliance networks (Chow et al., 2002b), but these networks are seldom designed for source apportionment. Compliance monitoring sites do not usually provide the contrast needed between different times of the day and proximity to or distance from suspected contributors. Filter substrates used for compliance are not usually compatible with all of the needed measurements. Chow and Watson (1994a) and Chow et al. (1996a) describe a "Level 1" (i.e., preliminary or first-level) particulate matter (PM) assessment that can be completed with existing filters as a pilot study to form a conceptual model. The inaccuracy of the Level 1 assessment usually precludes justification for advanced emissions reduction strategies. The following eight steps should be performed when conducting a source apportionment study.

(1) *Formulate conceptual model*: The conceptual model provides a plausible, though not necessarily accurate, explanation of the sources, their zones of influence, transport from distant areas, timing of emissions throughout the day, and meteorology that affects relative emissions rates, transport, dispersion, transformation, and receptor concentration. The conceptual model guides the location of monitoring sites, sampling periods, sampling frequencies, sample durations, the selection of samples for laboratory analysis, and the species that are quantified in those samples. A conceptual model can be postulated from studies in similar areas, from smaller pilot studies (e.g., analysis of archived filters), and analysis of existing air quality and meteorological data.

(2) *Compile emissions inventory*: Receptor models need to be supplied with sources that are potential contributors. A receptor model inventory requires only source categories, not the locations and emission rates of specific sources. While ducted point source emissions can be reasonably estimated through source tests and operating records, area and mobile source emissions are inexact. The most common cause of differences between relative source contributions from source and receptor models is inaccurate emissions estimates (Core et al., 1981; Ryan et al., 1988; Watson and Chow, 2000; Watson et al., 2000). Receptor model results focus resources on those source emissions that are the most important contributors to excessive PM concentrations.

(3) *Characterize source emissions*: Chemical or physical properties that are believed to distinguish among different source types are measured on representative emitters. Source profiles are the mass abundances (fraction of total mass) of 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. Several compilations of particle profiles have been produced that might be applicable to a Level 1 source assessment (Watson, 1979; Sheffield and Gordon, 1986; Cooper et al., 1988; Olmez et al., 1988; Ahuja et al., 1989; Chow and Watson, 1989, 1994b; Core, 1989; Houck et al., 1989, 1990; Shareef et al., 1989; Watson and Chow, 2001a; Watson et al., 2001b). These profiles do not necessarily represent those that affect an area where they were not measured.

(4) *Analyze ambient samples for mass, elements, ions, carbon, and other components from sources*: Elements, ions (chloride, nitrate, sulfate, ammonium, water-soluble sodium, and water-soluble potassium), and organic (OC) and elemental carbon (EC) are sufficient to account for most of the particle mass. Additional properties such as molecular organic compounds, isotopic abundances, and single particle characteristics further distinguish source contributions from each other, even though they may not constitute large mass fractions.

(5) *Confirm source types with multivariate model*: If a sufficient number of chemically characterized ambient samples is available (more than 50), PCA and other multivariate analyses are helpful to determine the source types and profile characteristics that might be contributors.

(6) *Quantify source contribution*: The CMB model estimates source contributions based on the degree to which source profiles can be combined to reproduce ambient concentrations. The CMB attributes primary particles to their source types and determines the chemical form of secondary aerosol when the appropriate chemical components have been measured. Modern CMB software (Watson et al., 1990b, 1997b) requires specification of input data uncertainty and calculates standard errors for source contribution estimates. Table 1 provides detailed references on how to use the CMB.

(7) *Estimate profile changes and limiting precursors*: Source characteristics may change during transport to the receptor, the most common being changes of sulfur dioxide and oxide of nitrogen gaseous emissions to sulfate and nitrate particles. These changes can be simulated with aerosol evolution models (Watson and Chow, 2002a). Secondary ammonium sulfate and ammonium nitrate involve ammonia from non-combustion sources that may be a limiting precursor. Chemical equilibrium receptor models determine the extent to which one

precursors need to be diminished to achieve reductions in ammonium nitrate levels.

(8) *Reconcile source contributions with other data analyses and source models*: Since no model, source or receptor, is a perfect representation 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 receptor models and emissions inventories or source model should be reconciled. A "weight of evidence" from multiple source attribution approaches should add confidence to the control strategies that are developed.

Examples are given below for each of the eight steps to be performed in a source apportionment study.

2.1. Step 1: Form a conceptual model

A conceptual model describes the relevant physical and chemical processes that affect emissions, transport, and transformation by taking advantage of the large body of scientific knowledge already acquired. Citations in Table 1 propose several conceptual models (e.g., Pun and Seigneur, 1999; Watson and Chow, 2002a) that might be adapted to other areas. Widely applicable conceptual models and their implications for receptor model monitoring are:

Motor vehicle exhaust is the major PM contributor and originates mostly from downtown traffic during morning rush hours. Carbon measurements are taken before, during, and after rush hours at downtown and residential locations. Specific organic markers for diesel and gasoline exhaust are quantified.

The majority of particulate sulfate forms by regional photochemical reactions. Sulfate measurements are taken at an urban site and at an upwind regional site during the summer and compared with similar sulfate measurements during winter.

The majority of personal exposure results from sidestream cigarette smoke. Nicotine measurements are acquired from personal samplers attached to representative individuals that spend time in smoking and non-smoking environments.

Non-urban windblown dust is a major source of urban particle concentrations. Elemental measurements are taken at a downtown site and in an up-wind barren area (e.g. desert, playa, unplanted field) during windy and non-windy situations. Short time duration (<5 min) particle measurements are acquired with a continuous monitor and correlated with wind speeds of similar duration.

Denver measurements were based on the following conceptual model for $PM_{2.5}$: (1) secondary ammonium nitrate is a large component of wintertime $PM_{2.5}$, is of local rather than regional origin, is dominated by oxides

of nitrogen emissions from mobile emissions rather than elevated power station emissions, and is limited by nitric acid production rather than by available ammonia; (2) PM carbon is dominated by diesel exhaust and wood smoke, with less important contributions from gasoline exhaust and cooking; and (3) fugitive dust contributions are minimal, except when roads are sanded after snow, and is from manmade deposits rather than natural wind erosion during winter. To evaluate these concepts, nitric acid and ammonia gas measurements were needed with particulate ammonium nitrate and ammonium sulfate measurements at urban and non-urban sites. Specific organic compounds were needed to separate the different carbon source contributions. Elemental concentrations were needed to differentiate coal fly ash from suspended dust. Urban and non-urban monitors were needed with nighttime and morning samples that would bracket morning traffic and nighttime heating emissions. Testing this conceptual model also required extensive meteorological measurements at the surface and aloft to determine how surface and elevated emissions would mix throughout the day.

2.2. Step 2: Compile emissions

Emission models (Dickson and Oliver, 1991) estimate temporal and spatial emission rates based on activity level, emission rate per unit of activity, and meteorology. Emissions models apply emission factors measured for a range of activity levels from representative sources to values for those activities estimated for a study area. Commonly used activity surrogates are: (1) fuel consumed, product produce, 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. Activity data are often unavailable for the spatial scales desired of source models, and emissions are estimated for a larger area (e.g., a county, state, or province) and spatially allocated to a smaller scale based on population census tracts, land-use maps, and roadway networks. An accurate inventory for source modeling often requires greater effort and expense than field monitoring for air quality concentrations and meteorology.

US EPA (1999) publishes AP-42 emission factors used to formulate national inventories, but these are often inappropriate for specific airsheds, especially those outside of the US. This is recognized in the AP-42 introduction that recommends locally derived emission factors, where possible, and reiterates the limitations of factors derived from a few tests at other times and places.

Emissions inventory models are often used to develop control strategies by linear rollback modeling (Barth, 1970; deNevers and Morris, 1975; Cass, 1981; Cass and McRae, 1981, 1983). The linear rollback model assumes that atmospheric concentrations in excess of background are proportional to aggregate emission rates. Reducing excessive concentrations of a pollutant to levels below a pre-set standard requires emissions reductions that are proportionally equal to the relative amount by which the standard is exceeded. Receptor models are often used in conjunction with linear rollback to determine the contribution of source categories to excessive concentrations. The linear rollback is then performed on a category-specific basis, starting with the largest contributors. This is a more accurate method of justifying emissions reductions than a rollback of unspeciated PM because the relative emissions from individual sources within a category are believed to be more accurate than the absolute emissions within the category or the relative emissions between source categories (e.g., Cass and McRae, 1981, 1983).

While source models need spatial and temporal resolution and very accurate emissions rates, receptor models need only a seasonal or annual average, area-wide inventory to identify potential source categories. Individual emitters must be grouped into more generalized categories with similar source profiles. For example, an outdoor inventory will often contain separate entries for power generation, industrial, and institutional coal combustion. Since these combustion processes, and often the coal, are similar in a given airshed, it is unlikely that their contributions can be distinguished by the CMB and they must be combined into a "coal-burning category". Source categories that are often combined for particulate receptor modeling are:

Vegetative burning and cooking: Fireplaces, wood stoves, prescribed burns, wildfires, char-broiling, and meat cooking. Some of these sub-categories may be separated when appropriate organic compounds are measured.

Diesel exhaust: Heavy and light duty cars and trucks, off-road equipment, stationary engines for pumps and generators, and locomotives.

Gasoline exhaust: Heavy and light duty cars and trucks, and small engines. Emissions inventories do not usually contain breakdowns by cold-starts and poorly maintained vehicles, although these might be discriminated by certain organic compounds in a profile. Since leaded fuels are no longer used in the US, there is no need to separately apportion use of this fuel, but leaded gasoline is still in use in many other countries.

Fugitive dust: Paved roads, unpaved roads, agricultural tilling, construction, wind erosion, and industrial aggregate. These can sometimes be divided into sub-categories based on single particle shape and elemental content or the measurement of specific mineral composition.

Metals: Copper smelters, lead smelters, steel mills, and aluminum mills. These often have similar metal emissions but in different abundances depending on the process.

Aggregate handling: Cement, quarrying, and mining. Ores, in particular, are often enriched in the materials being extracted and sub-categories may be defined for these cases. When low level measurements of trace elements such as copper, zinc, and lead are made, metal processing operations that use these materials can be classified into separate categories.

Indoor inventories might include the following particulate source types (Sexton et al., 1986; Sexton and Hayward, 1987; Koutrakis et al., 1992): (1) outdoor air that infiltrates through doors, windows, and poor insulation; (2) house dust from track-in and resuspension from vacuuming and personal movement; (3) cooking, both from the fuel combustion and the food, especially during frying; (4) sidestream smoke when the building contains smokers; (5) animal dander and fur in the presence of pets and other animals; (6) molds, spores, and fungi, especially those that form and are entrained from ventilation ducts; (7) vehicle exhaust in commuter transport compartments; and (8) a large variety of occupational emissions, depending on the type of work involved.

Table 2 summarizes wintertime outdoor emission rates for several source categories in the Denver, CO, metropolitan area. Similar sources and pollutants are found in most urban areas. Inventories should include primary particles that are directly emitted by sources (PM_{10} and $PM_{2.5}$) as well as gaseous precursors for

secondary particles that form in the atmosphere. Sulfur dioxide, ammonia, and oxides of nitrogen are the precursors for sulfuric acid, ammonium bisulfate, ammonium sulfate, and ammonium nitrate particles that often constitute major fractions of $PM_{2.5}$ and PM_{10}. Volatile organic compounds (VOC) are involved in photochemical reactions that engender sulfuric acid, nitric acid, particle-phase organics, as well as ozone. VOC precursors that convert to secondary organic particles are those with more than 6–8 carbon atoms (Pandis et al., 1992), and canister-based hydrocarbons are only valid for species with less than 12 carbon atoms.

Table 2 reflects the fact that most particle emissions from combustion sources are in the $PM_{2.5}$ fraction, while fugitive dust emissions are mostly in the coarse ($PM_{10} - PM_{2.5}$) fraction. This is consistent with source measurements of particle size fractions. Coarse particles are produced by grinding of larger particles to smaller particles while fine particles are produced by condensation, coagulation, and gas-to-particle conversion common to combustion sources.

Table 2 indicates that fugitive dust sources from paved and unpaved roads, sanding, and construction constitute 79% of PM_{10} and 43% of $PM_{2.5}$ emissions. Fugitive dust is a large fraction of all emissions inventories and is inconsistent with the amount of geological material found in ambient air in most locations. Detailed examination of fugitive dust emission factors (Watson and Chow, 2000; Watson et al., 2000; Countess et al., 2001) shows that these are derived from horizontal fluxes in upwind/downwind experiments in

Table 2
Wintertime emissions estimates for the Denver, CO, metropolitan area[a]

Source type	Source emission rate estimates (tons/day)						
	PM_{10}	$PM_{2.5}$	SO_2	NO_x	NH_3	VOC	CO
Gasoline exhaust	1.7	1.6	3.3	137.7	_[b]	157.6	1340.8
Visibly smoking gas exhaust	0.2	0.2	_[b]	_[b]	_[b]	_[b]	_[b]
Diesel exhaust	5.0	4.9	1.5	36.1	_[b]	8.4	30.9
Off-road diesel exhaust	1.8	1.8	1.7	27.4	_[b]	14.3	111.7
Wood burning	1.8	1.8	0.0	0.0	_[b]	_[b]	_[b]
Road dust and sand	49.6	7.4	0.0	0.0	_[b]	_[b]	_[b]
Coal power stations	1.3	0.7	62.1	64.3	_[b]	_[b]	_[b]
Other industries	7.8	2.6	16.7	47.8	_[b]	_[b]	_[b]
Natural gas	0.5	0.5	0.0	28.4	_[b]	_[b]	_[b]
Unpaved road dust	28.2	4.2	0.0	0.0	_[b]	_[b]	_[b]
Restaurant cooking	1.4	1.4	0.0	0.0	_[b]	_[b]	_[b]
Construction dust	2.2	0.3	0.0	0.0	_[b]	_[b]	_[b]
Biogenic	_[b]	_[b]	_[b]	3.0	_[b]	31.3	0.0
Industrial	_[b]	_[b]	_[b]		_[b]	34.8	22.3
Area sources	_[b]	_[b]	_[b]	1.0	_[b]	89.8	72.3
Total	101.6	27.5	85.2	345.7	_[b]	336.2	1578.0

[a] From Regional Air Quality Council (1998), ''Review of Blueprint for Clean Air Emissions Inventory'', April 8 as summarized in Watson et al. (1998b).
[b] Not estimated for the Denver area owing to lack of activity data or emissions factors.

which 60–90% of the emissions are within the lowest two meters above ground level. Since these low level emissions are likely to deposit within a few hundred meters of the emissions point, they should not be part of emission factors used to represent urban-scale inventories.

Mobile source combustion emissions from diesel and gasoline engines operating on- and off-road are important contributions in nearly all urban areas. Fujita et al. (1997) identified two-stroke engines, common on small motorbikes, as an important mobile source category in Bangkock, Thailand. For the Denver area, Table 2 shows that motor vehicle exhaust (the sum of gasoline, diesel, and off-road emissions) constitutes 31% of $PM_{2.5}$ emissions, with diesel (including off-road) representing more than 79% of vehicle exhaust. Poorly maintained gasoline vehicles, identified by visible exhaust, constitute 11% of gasoline-fueled vehicle emissions and only 2% of total vehicle emissions.

Vehicle emission testing is expensive, and it is difficult to recruit commercially used trucks for testing; as a result, the California emissions inventory is based on tests of 6000 light-duty gasoline vehicles, as compared to only 70 heavy-duty diesel trucks (Lloyd and Cackette, 2001a,b). Recent tests have shown large discrepancies between motor vehicle emission factors and on-road tests for a variety of pollutants. Hansen and Rosen (1990) report measurements from Berkeley, CA, of carbon emissions from individual vehicles in on-road operation. They measured the ratio of light-absorbing carbon to CO_2 as vehicles drove past the Lawrence Berkeley Laboratory, and found a factor of 250 between the highest and lowest ratio for 60 gasoline-fueled vehicles. They did not categorize vehicles by weight and age, but the range of ratios shows the potential for large uncertainties in these factors. Further evidence of these discrepancies was found by using remote sensing of individual tailpipe emissions for carbon monoxide, hydrocarbons, and oxides of nitrogen (Bishop et al., 1989, 1996, 1997; Ashbaugh et al., 1992; Stedman et al., 1993; Zhang et al., 1993; Cadle and Stephens, 1994; Stephens et al., 1996; Johnson et al., 1998; Singer et al., 1998). Remote sensing shows that approximately 20% of the on-road vehicles emit nearly 80% of the pollutants. Excess emissions are due to off-cycle operating conditions and poor-maintenance that are not well represented by laboratory dynamometer studies. Remote sensors for suspended particles are still being developed, although excessive gaseous emissions detected by current technology probably indicate poor combustion properties that also increase particle emissions over those currently estimated by emissions models.

Table 2 shows that residential wood combustion (RWC) constitutes ~7% of $PM_{2.5}$ emissions for wintertime Denver. This estimate includes the effectiveness of control measures such as new technology stoves and no-burn days that were implemented after a 1988 receptor modeling study found wood-burning contributing 25% of wintertime $PM_{2.5}$ in Denver (Watson et al., 1988). Special tests of wood-burning emissions (McDonald et al., 2000) using contemporary wood-burning methods were made for the Denver area.

The impact of cooking on indoor air has been well documented (Truesdale, 1982; Davidson et al., 1986; Reid et al., 1986; Ahuja et al., 1987; Raiyani et al., 1993; Smith, 1995; Ballard-Tremeer and Jawurek, 1996; Fan and Zhang, 1998; Albalak et al., 1999; Lee et al., 2001), and its effects on outdoor $PM_{2.5}$ concentrations have been identified (Hildemann et al., 1991; Rogge et al., 1991; McDonald et al., 1998; Kleeman et al., 1999; Nolte et al., 1999; Schauer et al., 1999; Mugica et al., 2001). Table 2 indicates that ~5% of $PM_{2.5}$ is emitted by this source. For Table 2 emissions, refineries, cement production, grain handling, and aggregate handling operations were quantified with 12% of $PM_{2.5}$ emissions; coal-fired power stations emitted 2.5% of $PM_{2.5}$ while other combustion, mostly with natural gas, is estimated to contribute ~2% of the total.

Table 2 shows nearly 60% of the oxides of nitrogen deriving from mobile sources, with coal-fired power generation emitting 19% of the total. Coal is the major sulfur dioxide emitter in areas where it is used, and this is evident for Denver. Motor vehicle fuels also include sulfur that may be more influential for the formation of wintertime sulfate if it is emitted into ground-level fogs under stagnant conditions where conversion can be very rapid. A significant quantity (8%) of sulfur dioxide derives from mobile sources, with half of that deriving from on-road and off-road diesel exhaust.

Fig. 1 shows how inventory $PM_{2.5}$ emissions are combined into source categories that can be distinguished at receptors by their chemical components. This figure assumes that measured organic compounds used in CMB calculations can distinguish among different carbon sub-types such as cooking/burning and gasoline/diesel exhaust. These sub-types usually cannot be distinguished by commonly applied elemental/ion/carbon analyses. All of the fugitive dust sources are combined because distinguishing species have not yet been developed that conclusively separate paved road, unpaved road, and construction sources. Salt used as a de-icing material can be detected, but it does not have a constant abundance relative to other geological materials because it is applied intermittently and is soon washed away by melting snow.

2.3. Step 3: Characterize sources

Analogous to the measurement of emission factors, source profiles are created by sampling emissions from a variety of single emitters or small groups of emit-
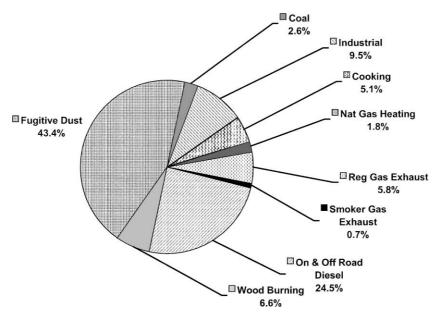
Fig. 1. PM$_{2.5}$ emissions from Table 2 are combined into categories that might be resolved by receptor models. Paved road dust, unpaved road dust, and construction emissions are summed, as are on-road and off-road diesel emissions.

ters. These samples are then submitted to a variety of chemical and physical analyses to determine those properties that will allow contributions from the sources they represent to be distinguished at receptors. The mass for each of these properties is normalized to a common property in the emissions from all sources, typically the PM$_{10}$ or PM$_{2.5}$ mass emissions rate. This mass fraction is termed the species abundance.

Individual profiles are formed from single samples, and the precisions of the numerator and denominator of the species abundances are propagated (Watson et al., 2001c) to obtain the individual profile uncertainties. These individual profiles are further composited to obtain the source profiles used for CMB source apportionment. The simplest composite consists of the average and standard deviation of species abundances for all individual profiles within a group. For example, if ten tests of diesel vehicle exhaust are taken, the composite species abundance is an average of the ten individual profile abundances and the uncertainty is the standard deviation of that average. Outlier tests are applied to reject individual profiles that unduly bias the average and standard deviation of the composite profile. There are always some outliers in any series of source tests, usually for reasons that can never be determined. It is important to obtain ten or more samples that run the range of operating conditions and fuels in an area to develop source profiles.

The ideal source sampling method would allow for chemical and physical transformations of source emissions to occur prior to sample collection. Lacking this ideal, the sampling would at least quantify the precursors of the receptor profile so that a theoretically or empirically derived transformation could be applied. The following methods (Gordon et al., 1984; Chow et al., 1988a; Watson and Chow, 2001a; Watson et al., 2001b) are commonly used for different source types to obtain samples from which profiles may be determined:

Hot exhaust sampling: Effluent is extracted from a duct or stack at emissions temperatures and drawn though filters. The EPA "Method 201" stack test method is most commonly applied in the US to determine compliance with PM$_{10}$ emissions standards. Hot exhaust does not permit the condensation of vapors into particles prior to sampling, and it sometimes interferes with the sampling substrate or container. In coal-fired station emissions, the selenium does not condense on other particles until temperatures approach ambient. Hot exhaust samples are not often taken on substrates or in containers amenable to extensive chemical analysis. Even though it is widely used for compliance, hot exhaust sampling is not appropriate for receptor modeling studies.

Diluted exhaust sampling: Effluent extracted from a duct is mixed with clean ambient air so that gases can condense on particles (Heinsohn et al., 1980; Hildemann et al., 1989; Zielinska et al., 1998; England et al., 2000). The near-ambient temperature effluent is then drawn through substrates that are analyzed for the desired properties. Diluted exhaust samplers are used for laboratory simulations of emissions from individual sources. Dynamometer simulations use diluted exhaust sampling to estimate emissions for different vehicle types, fuels,

and driving conditions. Wood stove, fireplace, and cookstove emissions can also be simulated by dilution sampling of representatives in a laboratory or field environment.

Airborne sampling: Effluent is drawn from a plume aloft after it has cooled to near ambient temperatures but before it is dominated by the particles present in the background air. Aircraft, balloons, and cranes have been used to elevate sampling systems into the plume (Anderson et al., 1977, 1998; Armstrong et al., 1981; Hering et al., 1981; Gillani and Wilson, 1983; Shah et al., 1989; Cahill et al., 1992; Baxter and Pederson, 1994; Lagomarsino and Latner, 1997; Hofmann et al., 1998; Greenberg et al., 1999). With airborne sampling it is possible to follow a large plume and examine how source profiles change as secondary aerosol is formed. Difficulties of airborne plume sampling are: (1) locating the sampler in the plume instead of ambient air; (2) staying in the plume long enough to obtain a sufficient sample for chemical analysis; and (3) mixing of ambient air with the plume, so the source profile is really a combination of emissions and ambient air.

Ground-based sampling: Ambient samples are taken in locations and during time periods for which a single source type dominates the emissions. Ground-based source sampling methods are identical to receptor sampling methods with the requirements that: (1) meteorological conditions and sampling times are conducive to domination by a particular source; (2) samples are of short enough duration to take advantage of those conditions; and (3) aerosol from other interfering sources is low or can be apportioned and removed from the sample. Tunnels, parking garages, vehicle staging areas, and isolated but heavily traveled roadways are often used to obtain samples for motor vehicle exhaust. Tunnels are especially useful for this because a large number of vehicles can be evaluated with little interference from sources other than suspended road dust (Pierson and Brachaczek, 1976, 1983; Chang et al., 1981; Hering et al., 1984; Miguel, 1984; Lonneman et al., 1986; Benner et al., 1989; Dannecker et al., 1990; Pierson et al., 1990, 1996; Zielinska and Fung, 1994; Khalili et al., 1995; Barrefors, 1996; Bishop et al., 1996; Duffy and Nelson, 1996; Gertler and Pierson, 1996; Moeckli et al., 1996; Gertler et al., 1997; Weingartner et al., 1997; Fraser et al., 1998; Gillies et al., 1998, 2001; Rogak et al., 1998; Staehelin et al., 1998). Using short-duration source-dominated samples, Rheingrover and Gordon (1988) and Annegarn et al. (1992) characterized several point sources using ambient sampling downwind of the source. Chow (1985) examined the effects of an elevated coal-fired power plant emission on ground-based samples in a rural environment with this method.

Grab sampling and laboratory resuspension: A sample of pollution residue is obtained and suspended in a chamber for sampling onto filters (Chow et al., 1994; Carvacho et al., 1996). This is most applicable to non-ducted fugitive and industrial dust emissions. A sample swept, shoveled, or vacuumed from a storage pile, transfer system, or roadbed can be taken to represent these source types. Five to ten different samples from the same source are averaged to obtain a representative source profile.

Ground-based and grab sampling are the most cost-effective and practical methods for most situations, although large industrial stack emissions require diluted sampling and mobile source sub-types (e.g., high emitting vehicles) can only be isolated in laboratory dynamometer tests.

Source profiles for Denver are qualitatively similar to those found elsewhere, although the abundances vary depending on local conditions. In geological material, aluminum (Al), silicon (Si), potassium (K), calcium (Ca), and iron (Fe) have large abundances with low variabilities. The total potassium (K) abundance is often 10–30 times the abundance of soluble potassium (K^+) in fugitive dust. Aluminum (Al), potassium (K), calcium (Ca), and iron (Fe) abundances are similar among dust profiles. Lead (Pb) is sometimes abundant in paved road dust, but it is <0.005% in the other geological profiles, probably due to deposition from previously emitted leaded-gasoline vehicle exhaust or remnants of lead from the exhaust trains of older vehicles. EC abundances are highly variable in geological material, and are often negligible in natural soil samples. OC is typically 5–15% in geological emitters. OC is most abundant in paved road and agricultural dusts, although the specific compounds probably differ for these two sources. Vehicle brake wear, tire wear, and oil drips could result in greater abundances of Pb, EC, and OC in paved road dust. Soluble sulfate, nitrate, and ammonium abundances are usually low, in the range of 0–0.5%. Sodium and chloride are also low, with less than 0.5% in abundance. Larger abundances of these materials may occur soon after roadway de-icing.

OC and EC are the most abundant species in motor vehicle exhaust, typically accounting for over 95% of the total mass. 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. Watson et al. (2001b) found an OC/TC ratio of 0.58 in the composite vehicle profile for northwestern Colorado. Earlier measurements in Denver, CO (Watson et al., 1990a) reported an OC/TC ratio of 0.39 for the cold transient cycle and 0.81 for the cold stabilized cycle.

OC and EC are also abundant in wood burning and cooking emissions. Watson et al. (2001b) compared RWC, residential coal combustion (RCC), and forest fire $PM_{2.5}$ profiles. Average OC abundances ranged from ~50% in RWC and the forest fire profiles to ~70% in the

RCC profile. EC averaged 3% in forest fire, 12% in RWC, and 26% in RCC. The OC/TC ratio was highest in the forest fire profile (0.94) and similar for the two residential combustion profiles, with 0.73 in RCC and 0.81 in RWC. Watson and Chow (2001a) measured profiles for asparagus field burning in California's Imperial Valley with OC/TC ratios of 0.93, similar to the 0.94 ratio for forest fire emissions. A similar observation was made for charbroil cooking emissions, with 60–70% OC abundances and high (>0.95) OC/TC ratios. The K^+/K 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 combustion conditions and different emissions. Sulfate is one of the most abundant constituents in the particle phase and sulfur dioxide 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 when it has not reacted or deposited significantly during transport to a receptor. Crystal elements such as Si, Ca, and Fe are often present at 30–50% of the corresponding levels in geological material, while Al abundances are similar to or enriched over those found in surface soils. Other elements such as phosphorus (P), potassium (K), titanium (Ti), chromium (Cr), manganese (Mn), strontium (Sr), zirconium (Zr), and barium (Ba) are often detectable at less than 1% levels.

All of the elemental abundances are highly dependent on the ash composition of the parent coal, as well as the specific combustion process and pollution control devices. Watson et al. (2001b) detected selenium (Se) at the level of 0.2–0.4% in coal-fired power station emissions with no scrubbers or wet scrubbers, but not in emissions from a unit with a dry limestone scrubber. Se is usually in the gaseous phase within hot stack emissions, and it condenses on particles when air is cooled in the dilution chamber. Abundances of Ca (15%), Cl (1%), and nitrate (1%) in the limestone-scrubbed unit were a few times higher than in the other units. These differences may have resulted from the dry lime scrubber, which added some calcium and absorbed the selenium in the vapor phase.

These examples show that although there are similarities in chemical compositions for different sources, using source profiles from one airshed or time period may not provide a valid CMB apportionment for ambient samples in another airshed or in another time period. Source emissions of precursor gases 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. Trace metals acquired from elemental analysis

of Teflon-membrane filters are only abundant in the geological and some industrial profiles. Elemental measurements by themselves are necessary, but insufficient, for a receptor modeling study. Chemical speciation must also include ammonium, sulfate, nitrate, OC, and EC. Separating carbon fractions with the use of different combustion temperatures and atmospheres can also be a useful tool in source apportionment studies (Chow et al., 1993b). Watson et al. (1994a), using the thermal evolution method in the IMPROVE protocol (Chow et al., 1993b, 2001), showed that gasoline- and diesel-powered vehicle emissions exhibit different abundances of different fractions of OC and EC. Engelbrecht et al. (2002) used seven carbon fractions in their CMB analysis to distinguish source contributions between regular and low-smoke RCC. Simultaneous gas measurements as well as other characteristics of suspended particles will be needed as more refined control strategies are developed using receptor models.

The Denver example measured organic compounds such as those listed in Table 3 in both source and receptor samples to apportion source sub-types such as meat cooking, wood burning, and exhaust from different vehicle operating modes. Sampling and analysis methods for organic compounds are still being perfected. Organic analyses are more difficult or costly to apply than currently available elemental, ion, and carbon measurements.

2.4. Step 4: Analyze samples for material balance

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, de-icing, or windblown dry lake beds). Several summaries of particle sampling and analysis methods (Watson and Chow, 1993, 1994, 2001b; Chow and Watson, 1994a, 1998; Chow, 1995) describe ambient sampling and chemical analysis options. The material balance requires elemental, ionic, and carbon analyses. Although not a source apportionment, the material balance provides guidance on which components are the major cause of excessive mass concentrations. This knowledge can focus efforts to improve the emissions inventory for source modeling and allows a speciated linear rollback model to be applied for initial control strategy development. The following sample analysis hierarchy (updated from Chow et al., 1996a) demonstrates how elemental, ionic, and carbon concentrations can be measured for different levels of resources:

Analyze archived filter samples: High-volume quartz-fiber filter samples used to determine mass for compliance with air quality standards are sub-optimal for chemical analysis, but they are commonly available throughout the world. These filters have already been

Table 3
Organic compounds from different OC emission and in ambient air

Species	Predominant sources	Particle–gas phase distribution
PAH		
Naphthalene	Motor vehicles, wood smoke	Gas phase
Methylnaphthalenes	Motor vehicles, wood smoke	Gas phase
Dimethynaphthalenes	Motor vehicles, wood smoke	Gas phase
Biphenyl	Motor vehicles, wood smoke	Gas phase
Acenaphthylene	Motor vehicles, wood smoke	Gas phase
Acenaphthene	Motor vehicles, wood smoke	Gas phase
Fluorene	Motor vehicles, wood smoke	Gas phase
Phenanthrene	Motor vehicles, wood smoke	Particle–gas phase
Anthracene	Motor vehicles, wood smoke	Particle–gas phase
Fluoranthene	Motor vehicles, wood smoke	Particle–gas phase
Pyrene	Motor vehicles, wood smoke	Particle–gas phase
Retene	Motor vehicles, wood smoke	Particle–gas phase
Benzo[*b*]naphtho[2,1]thiophene	Motor vehicles	Particle phase
Benz[*a*]anthracene	Motor vehicles, wood smoke	Particle phase
Chrysene	Motor vehicles, wood smoke	Particle phase
Benzo[*b + j + k*]fluoranthene	Motor vehicles, wood smoke	Particle phase
Benzo[a]pyrene	Motor vehicles, wood smoke	Particle phase
Indene[123-*cd*]pyrene	Motor vehicles, wood smoke	Particle phase
Dibenzo[*ah + ac*]anthracene	Motor vehicles, wood smoke	Particle phase
Benzo[*ghi*]perylene	Motor vehicles, wood smoke	Particle phase
Coronene	Motor vehicles, wood smoke	Particle phase
Hopanes and Steranes		
Cholestanes	Motor vehicles	Particle phase
Trisnorhopanes	Motor vehicles	Particle phase
Norhopanes	Motor vehicles	Particle phase
Hopanes	Motor vehicles	Particle phase
Guaiacols		
4-Methylguaiacol	Wood smoke	Gas phase
4-Allylguaiacol	Wood smoke	Particle–gas phase
Isouegenol	Wood smoke	Particle–gas phase
Acetovanillone	Wood smoke	Particle phase
Syringols		
Syringol	Wood smoke, mostly hardwood	Particle–gas phase
4-Methylsyringol	Wood smoke, mostly hardwood	Particle–gas phase
Syringaldehyde	Wood smoke, mostly hardwood	Particle phase
Lactones		
Caprolactone	Meat cooking	Gas phase
Decanolactone	Meat cooking	Particle–gas phase
Undecanoic-γ-lactone	Meat cooking	Particle–gas phase
Sterols		
Cholesterol	Meat cooking	Particle phase
Sitosterol	Meat cooking, wood smoke	Particle phase

weighed to determine mass concentration. They can be analyzed for elements by X-ray fluorescence (Watson et al., 1999a) with appropriate corrections for filter absorption and blank subtraction; for ions by ion chromatography (Chow and Watson, 1999) and atomic absorption spectrophotometry (Chow et al., 2002b), and for carbon by thermal combustion carbon analysis (Chow et al., 1993b, 2001). Aluminum and silicon cannot be quantified on these samples because they are part of the filter matrix. Other trace elements may also be present in the filter matrix as impurities, and large fractions of volatile species such as ammonium nitrate may have evaporated during storage (Witz et al., 1990). For $PM_{2.5}$ standard attainment, US EPA requires sampling with low-volume federal reference method (FRM) samplers equipped with Teflon-membrane filters. These samples can be analyzed for elements by X-ray fluorescence. Sulfate can be estimated from the sulfur concen-

tration. Nitrate and sulfate ions can be subsequently measured by ion chromatography, except that heating and vacuum stages during X-ray fluorescence analysis enhances nitrate volatilization and results in a low estimate of nitrate concentration.

Planned compliance sampling: If it is known that chemical analyses will be applied to some or all of the high-volume PM_{10} or low-volume $PM_{2.5}$ samples, several precautions should be taken. When procuring filters, minimally acceptable blank concentrations should be stated for each chemical to be quantified. Filter holders that mate to the high-volume PM_{10} sampler or $PM_{2.5}$ FRM sampler should be obtained, and filters should be loaded and unloaded using gloved hands in a laboratory setting to minimize contamination. The passive period in the sampler before and after air is drawn through the filter should be minimized, and filters should be stored in clean containers under refrigeration after weighing.

Collocated filter sampling: An additional sampler with a Teflon-membrane filter appropriate for complete elemental analysis can be collocated with the high-volume PM_{10} compliance sampler. This filter should also be pre- and post-weighed and submitted to the same acceptance testing and storage conditions described for planned compliance sampling. For the $PM_{2.5}$ FRM, collocated quartz-fiber filters appropriate for ion and carbon analysis are needed. Chow (1995) identifies several sampling systems that can be collocated. A Minivol sampler with a 5 l min^{-1} flow rate (Baldauf et al., 2001; Chow et al., 2002a) is a cost-effective method of adding additional filter media to a sampling site.

Collocated continuous monitor sampling: Watson et al. (1998a) and McMurry (2000) describe several continuous particle detectors that can obtain mass, mass-surrogates, and chemical- or size-specific concentrations over averaging periods of 1–60 min duration. These monitors can identify diurnal patterns, associate high concentrations with wind directions from sources, and provide indicators of concentrations between infrequent filter samples (which are often every sixth day). Nephelometers with "smart" heaters are the most cost-effective continuous monitors. These measure light scattering rather than particle mass, but good relationships between the two variables can often be established (Chow et al., 2002c). Since light scattering increases rapidly as soluble particles take on water at humidities exceeding 70%, a smart heater in the sample line provides a temperature increase sufficient to maintain sampler humidities below this threshold. Constant heating at high temperatures (typically >30 °C) causes volatile particles to evaporate, thereby negatively biasing the mass estimate.

Sequential filter sampling: Daily samples, or samples taken during different parts of the day, are often needed to evaluate pollution buildups or to relate concentra-

tions to human exposure or observed health end-points. Sequential filter samplers using several filter media in parallel have been used to acquire these types of filters for subsequent analysis (e.g., Chow et al., 1993a; Chow, 1995). Sequential sampling systems have been applied in $PM_{2.5}$ FRM networks across the US. Several filters are loaded and a timing mechanism switches the flow through different sets of filters at pre-set intervals.

Saturation sampling: An area can be saturated with small, battery-powered samplers to evaluate the zone of representation for long-term monitors and the zone of influence for source emissions. This strategy is useful for spatial correlation receptor models and when chemical profiles of specific emitters are too similar to differentiate them from each other. Samplers are located within and around the suspected emitters (Tropp et al., 1998; Blanchard et al., 1999; Chow et al., 1999, 2002a; Gillies et al., 1999).

Denuder and absorbant sampling: Volatile substances such as ammonium nitrate and organic compounds require denuders and gas-absorbing backup filters (Kitto and Colbeck, 1999). Precursor gases for chemical receptor models can also be obtained from these sampling strategies.

Fig. 2 illustrates a material balance for a single high volume TSP sample from a sample in Xian, China. This sample had a relatively high TSP mass loading (336 µg m^{-3}) and was measured on a day with stagnant meteorology in a residential area. The commonly held conceptual model in Xian is that most of the high TSP concentrations result from dust storms in the region
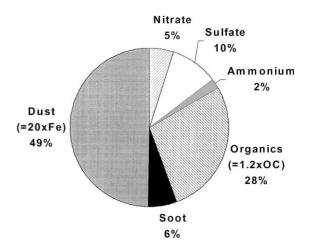
Fig. 2. Material balance for a 24-h 336 µg m^{-3} TSP quartz filter sample from an eastern urban site in Xian, China for October 27, 1997. Aluminum and silicon cannot be quantified on these samples because they are in the filter matrix and much of the nitrate may have volatilized before analysis. Fugitive dust emissions are estimated as 20 times the iron concentration and OC is multiplied by 1.2 to account for unmeasured hydrogen and oxygen associated with urban organic compounds.

(Zhang et al., 1996, 1997, 1998b). The simple material balance in Fig. 2 demonstrates that only half of the TSP is due to dust. Given the low wind speeds during the sampling period, most of this is probably from nearby roads and vacant land rather than windblown loess in the desert. This could be further verified by comparison with simultaneous samples taken at other sites closer to cleaner or dirtier roadways. OC and EC account for 34% of TSP mass, indicating that combustion sources are a major portion of TSP. Secondary sulfate constitutes a relatively low fraction. Although the nitrate concentration is a minor portion of TSP, it is probable that much of the original nitrate sampled was lost from the filter prior to chemical analysis. Additional archived samples for different time periods and locations would need to be analyzed and compared to form a conceptual model. Some of these should include periods of high winds

when desert dust might be a large contributor. This simple analysis on archived filters helps to focus emissions characterization efforts and future network design by establishing the most abundant components of the suspended particle mass. It suggests that primary emissions reductions for local dust and combustion sources (both mobile and area) should be pursued to reduce ambient concentrations.

The other extreme for the material balance is illustrated in Fig. 3 for two $PM_{2.5}$ pollution episodes near Denver, CO. Samples were taken during the morning, afternoon, and overnight on consecutive days. The days selected for detailed chemical analysis were determined after examining mass concentrations measured on each sample, thereby reducing the study cost by minimizing analyses on low concentration samples. Three pollution episodes can be detected in these data, each with dif-
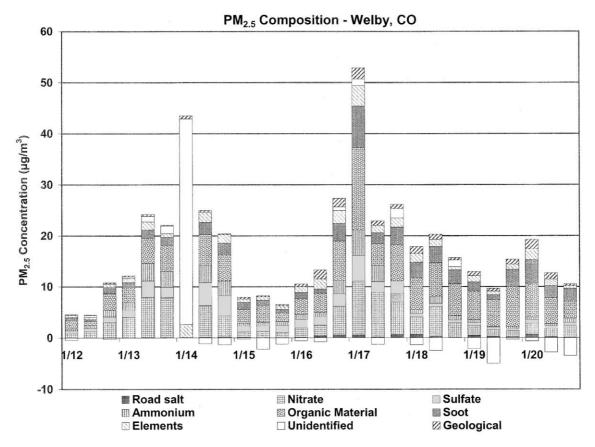

Fig. 3. Material balance for $PM_{2.5}$ concentrations measured at Welby, CO, located on the northern edge of Denver, resulting from gravimetric, elemental, ion, and carbon analysis (Watson et al., 1998b). Three samples per day were acquired from 0600 to 1200, 1200 to 1800, and 1800 to 0600 MST with the first sample centered over the date in 1996. Organic material is 1.4 times the OC measurement to compensate for unmeasured hydrogen and oxygen (Turpin and Lim, 2001). Geological is defined as $1.89Al + 2.14Si + 1.4Ca + 1.43Fe$ to account for unmeasured oxides in minerals. Soot is operationally defined as EC reported by the IMPROVE thermal/optical reflectance method (Chow et al., 1993b, 2001). The unidentified fraction is the difference between measured mass and the sum of other components. This is negative when the sum exceeds the measured mass. The unidentified amount is typically within measurement uncertainties when elements, ions, and carbon are measured.

ferent compositions. The first two that peak on the mornings of January 14, 1996 and January 17, 1996 have secondary ammonium nitrate and OC and EC as their most abundant components. The episode peaking on the morning of January 20, 1996 is dominated by OC and EC. Examining wind directions in conjunction with the evolution of these episodes shows $PM_{2.5}$ levels are highest when secondary sulfate and nitrate are transported back into the city where they mix with fresh emissions.

The morning sample on January 14, 1996 experienced an invalid sample for the quartz filter, so carbon and ion analyses were not performed. This sample illustrates the need for ions and carbon to obtain a material balance. The elements account for less than 10% of the measured $PM_{2.5}$ mass, even when the geological marker elements are weighted for unmeasured oxides.

2.5. Step 5: Multivariate analysis

Several multivariate data analysis methods should be applied to the data to confirm potential source contributions. These should make use of whatever air quality and meteorological measurements are available within the study area. Several useful analyses are described below.

Time series plots: Mass concentrations are plotted for each sampling period, similar to that illustrated in Fig. 3, and compared among different monitoring locations. Similar temporal patterns and concentrations indicate urban or regional scale source contributions, while a large divergence at one site indicates a middle-scale or neighborhood-scale source influence. This is extremely useful in short duration, continuous measurements. Using a successive moving average subtraction method, Watson and Chow (2001a) demonstrated that short-duration pulses in 5-min black carbon concentrations caused by nearby sources can be used in urban areas to estimate spatial zones of influence. Seasons and days of the week on which highest concentrations occur can be detected. Examination of these plots and comparisons among different sampling sites assists the selection of a sample subset for chemical analysis.

Diurnal plots: When continuous particle monitoring data are available, average concentrations can be calculated for each hour over a subset of days to identify patterns. These patterns can be associated with meteorological conditions and emissions for different sites. These plots often reveal increases in concentrations during morning and evening rush hours on weekdays at downtown locations that are not seen on weekends or at residential sites. This indicates that local traffic emissions are large contributors. The evening traffic peak may not appear during summer when mixed layers are deep later into the evening, but it will be very evident during winter when sunset corresponds to evening traf-

fic. Residential heating with wood or coal often appears as an elevated nighttime concentration that does not decay until very early the following morning.

Pollution roses: Averages of hourly or shorter time duration data can be calculated for wind direction sectors. Watson and Chow (2002a) showed that large concentrations associated with a given wind direction indicate a major source from that direction. Frequent wind direction shifts over a 24-h period usually preclude this type of analysis for day-long samples.

Time series correlation: Correlation coefficients can be calculated for chemical concentrations to determine which ones vary together and which ones do not. High correlation coefficients usually indicate origin from a common source or effects of common meteorology on all sources.

Spatial correlation: High spatial correlations among chemical components indicate urban or regional source influences, while low correlations indicate that the chemical component is mostly from nearby origin.

Eigenvector analysis: As explained in Table 1, time series or spatial eigenvector analysis in the form of PCA, FA, or EOF defines a smaller sample space for the measured chemical concentrations that can sometimes be related to source contributions. Source profiles from direct measurements are needed to interpret these eigenvectors.

All of these methods except the spatial eigenvector analysis were applied by Watson et al. (1998b) for the Denver study prior to estimating source contributions. These analyses provided the conceptual bases for selecting the source profiles applied to each individual sample on which chemical components were measured.

2.6. Step 6: Calculate source contributions

All of the multivariate models include, implicitly or explicitly, a CMB that quantifies source contributions. The CMB 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. Species 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 to the total mass and each chemical species.

The CMB modeling procedure requires: (1) identification of the contributing sources 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. Several solutions methods have been proposed for the CMB equations that are documented in Table 1 citations. Of these, 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, as documented in the theory and testing references of Table 1. Deviations from assumptions may increase the stated uncertainties of the source contribution estimates.

The formalized protocol for CMB model application and validation (Pace and Watson, 1987; Watson et al., 1991, 1998c) is applicable to the apportionment of gaseous organic compounds and particles. This seven-step protocol: (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 of 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
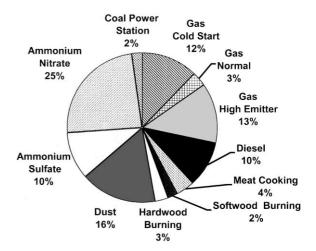

Fig. 4. Average 1997 wintertime source contribution estimates to PM$_{2.5}$ determined by the CMB receptor model for samples taken at Welby, CO, located on the northern edge of Denver, CO.

assessment methods. Chow and Watson (2002) found that this protocol was seldom followed in more than 20 recent CMB applications.

Fig. 4 shows average source contributions for the Denver samples obtained by following this protocol. Using organic compounds measured at source and receptor, it was demonstrated that meat cooking could be distinguished from wood burning. The three classes of gasoline vehicle exhaust (hot stabilized, cold start, and high emitters) could be distinguished by CMB from their source profiles. These sub-groups needed to be combined for receptor sites where only the elements, ions, and carbon were measured.

2.7. Step 7: Evaluate limiting precursors of secondary aerosol

Most applications use source profiles measured at the source, with at most dilution to ambient temperatures and <1 min of aging prior to collection to allow for condensation and rapid transformation. Profiles have been "aged" with aerosol evolution models (Watson et al., 2002), prior to submission to the CMB. These models simulate changes between source and receptor; they are often overly simplified and require additional assumptions regarding chemical mechanisms, relative transformation and deposition rates, mixing volumes, and transport times.

Changes in ammonia or nitric acid precursor concentrations do not proportionally result in reductions in ammonium nitrate concentrations. Ammonia concentrations might increase with emissions (e.g., larger fertilizer applications, a larger number of farm animals) or from reductions in sulfur dioxide emissions that would
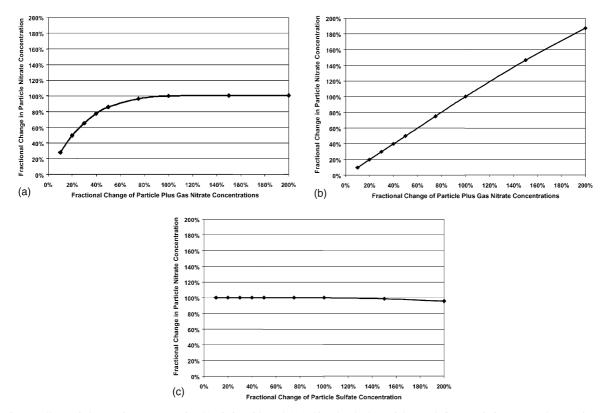
Fig. 5. Effects of changes in (a) ammonia, (b) nitric acid, and (c) sulfate levels that might result from emissions control strategies on average ammonium nitrate concentrations estimated using the simulating composition of atmospheric particles at equilibrium (SCAPE) method (Kim et al., 1993a,b; Kim and Seinfeld, 1995). SCAPE, an example of an aerosol equilibrium receptor model, apportions sodium, nitrate, sulfate, ammonium, and chloride among gas, liquid, and solid phases using thermodynamic equilibrium theory. These averages were derived from thousands of individual SCAPE simulations applied to 3-h periods over which particulate nitrate, sulfate, and ammonium and gaseous nitric acid and ammonia were measured at urban and non-urban receptor sites in the Denver area. Ambient temperature and relative humidity corresponding to the samples are the other SCAPE inputs. The horizontal axis represents the fraction of the measured 1997 concentration.

provide less sulfuric acid to react with existing ammonia. Conversely, ammonia emission reductions owing to urbanization might reduce ammonia concentrations. Ansari and Pandis (1998) and Blanchard et al. (2000) have created nomographs that allow a screening process for limiting precursors.

Fig. 5a–c shows how change in ammonia, nitric acid, and sulfate affect ambient concentrations in wintertime Denver. Fig. 5a shows no increases in ammonium nitrate result from increases in ammonia concentrations; a 25% reduction in ammonia results in a minor reduction in particle nitrate and a 50% ammonia reduction reduces particle nitrate by only 15%. After a 50% ammonia reduction, however, particulate nitrate decreases are nearly proportional to ammonia reductions. Ammonia levels must be reduced by more than half before large reductions of particulate nitrate are realized.

This contrasts with Fig. 5b that examines effects of nitric acid reductions on particulate nitrate. Change in

nitric acid, presumably resulting from changes in NO_x emissions that contribute to nitric acid, result in a direct and proportional reduction in $PM_{2.5}$ ammonium nitrate concentrations. Fig. 5c shows that there is no sensitivity to changes in sulfate levels. There is sufficient free ammonia in the Denver area that the small amount of ammonia freed by removing ammonium sulfate from the atmosphere has no effect on ammonium nitrate levels. Sulfate concentrations would need to increase by more than double to engender even a slight decrease in ammonium nitrate concentrations.

2.8. Step 8: Reconcile source contributions

Fig. 4 shows discrepancies with the proportional contributions from the inventory in Fig. 1. Comparisons between emissions inventories and receptor source contributions are not exact because: (1) primary particle inventories do not include the secondary aerosol
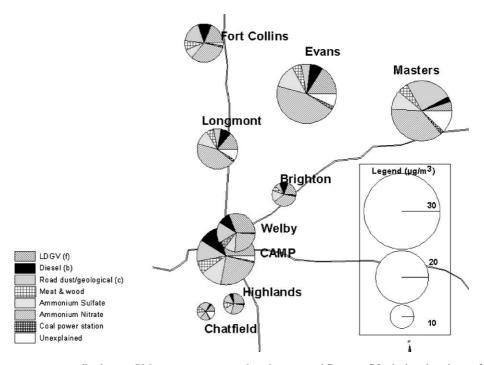
Fig. 6. Average source contributions to PM$_{2.5}$ at seven receptor locations around Denver, CO, during the winter of 1995–1996.

determined by receptor models; and (2) inventories are usually averaged over large areas and time periods while receptor model source contributions apply to the zone of representation of the receptor and to the time period over which samples were taken.

Major discrepancies were found between emissions estimates and receptor source apportionment of ambient samples for wintertime Denver, CO. Secondary ammonium sulfate and ammonium nitrate accounted for 35% of PM$_{2.5}$ in the receptor samples. Although sulfur dioxide and oxides of nitrogen emissions are often included in urban inventories, the fraction that converts to sulfate and nitrate can only be determined by source or receptor modeling. Ammonia emissions that engender the ammonium portion of these secondary contributions are seldom estimated. The emissions inventory estimated that 45% of emissions resulted from fugitive dust sources, while the receptor model source apportionment found the dust contribution to be 16%. The inventory showed that diesel exhaust constituted 80% of mobile source emissions, while receptor analysis with organic compounds showed that diesel exhaust constituted only ∼25% of the mobile source contributions. This finding has resulted in focusing efforts on better representing the emissions from gasoline vehicles in the inventory used for control strategy development and source modeling. This was a substantial deviation from the conceptual model stated earlier, and it is justification for refining that model.

Fig. 6 shows how source contributions vary among the seven measurement locations in the Denver area. There were similar PM$_{2.5}$ mass concentrations at both urban (Welby, CAMP, Fort Collins) and non-urban (Brighton, Evans, and Masters) sites, but the composition was quite different. The non-urban sites were dominated by secondary sulfate and nitrate. Analyzing sequential and diurnal samples along with wind flow patterns shows that oxides of nitrogen generated in the urban area mix in low lying fogs with ammonia from the rural areas results in higher secondary sulfate and nitrate levels that add mass to the primary particles generated within the city. This is another refinement to the original conceptual model.

3. Summary and conclusions

Receptor models complement source models for evaluating the contributions from different sources to measured concentrations. They use chemical characteristics measured at both source and receptor to accomplish this apportionment. Elemental analysis is necessary, but not sufficient for receptor models. Elements, ions, and carbon must be measured at a minimum. Additional chemical components such as organic compounds, carbon fractions, isotopic abundances, and single particle characteristics are methods that are being developed to further distinguish among source types.

Substantial information can be obtained from relatively inexpensive chemical analyses of archived filters, although these are not optimal for quantitative receptor source apportionment. The cost of a receptor modeling study needs to be balanced against the value of the information obtained from the study. Examples from a modern receptor study in wintertime Denver showed that emission estimates from gasoline vehicles were substantially underestimated and fugitive dust emissions were overestimated. The planned emissions reduction strategies to focus on diesel emissions reductions would not yield the expected benefits. Gasoline vehicle exhaust from cold starts and high emitting (poorly maintained) vehicles were identified as large contributors that were not adequately represented by the inventory. The intended control strategy of reducing oxides of nitrogen emissions to improve ammonium nitrate levels was confirmed by demonstrating that there is sufficient ammonia to neutralize available nitric acid. The costs of implementing emissions reduction strategies that did not result in the desired effects on air quality would be hundreds of times the cost of the receptor modeling study and would prolong human exposure in the area to excessive particle concentrations until the effective control strategies were identified.

Disclaimer

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