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Review of PM_{2.5} and PM₁₀ Apportionment for Fossil Fuel Combustion and Other Sources by the Chemical Mass Balance Receptor Model

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This review examines how the Chemical Mass Balance (CMB) receptor model has been used to quantify source contributions from fossil fuel combustion and other sources to ambient concentrations of PM_{2.5} and PM₁₀ for urban and regional scales. Nonfossil fuel sources, such as fugitive dust, cooking, vegetative burning, and natural or human-caused biogenics must be considered together with fossil-fuel sources in a CMB analysis to obtain closure for PM_{2.5} and PM₁₀ mass. CMB analyses in 22 different studies have found fossil fuel combustion to be a large contributor to PM_{2.5} and PM₁₀ concentrations, with most of the primary contributions originating from diesel- and gasoline-powered vehicle exhaust. Primary contributions from ducted sources, such as coal- and oil-fired power stations, are negligible when these facilities have been modernized with effective pollution controls, but they have been shown to be large contributors without these controls. Secondary sulfates and nitrates from fossil fuel combustion are recognized, but their attribution to specific precursor gas emitters is uncertain using either the CMB or source-oriented chemical transport models. Using source and receptor models together improves source contribution estimates and the confidence in those estimates.

Introduction

Fossil fuel combustion results in large quantities of primary particle emissions to the atmosphere, as well as gaseous compounds that convert to particles within hours to days after emission. Coal, raw and refined petroleum products, and natural gas are burned throughout the world to create electricity, refine metals and organic materials, heat buildings, cook food, and provide transportation. Fossil fuel combustion is also a major emitter of sulfur dioxide (SO₂) and oxides of nitrogen (NO_x), gases that react with other constituents in the atmosphere to create secondary sulfate and nitrate particles.¹ Much of the sulfate found in the air of the eastern United States,² in national parks with poor visibility,³ in waterways and snowpacks with high acidity,⁴ and in remote regions of Asia⁵ and Europe⁶ is believed to arise from fossil fuel sulfur emissions, especially coal combustion. In addition to direct emissions from stacks, chimneys, and tailpipes, fossil fuels

also result in fugitive dust from mining and extraction activities as well as disposal of noncombustible ash and pollution control residues that remain after combustion.

Effective methods have been developed and applied that use improved fuels, better combustion methods, and after-combustor treatment to reduce primary particles and precursor gases from fossil fuel combustion.^{7–9} These technologies are often costly and technologically challenging to implement; their initial applications need to be targeted at those emitters that have the highest contributions to ambient concentrations, especially in areas that exceed PM_{2.5} and PM₁₀ (mass of particles with aerodynamic diameters <2.5 and 10 μm, respectively) National Ambient Air Quality Standards (NAAQS).¹⁰ This targeting will become more important as additional and more costly emissions reduction measures are implemented to achieve natural visibility levels in U.S. national parks and wilderness areas required by regional haze regulations.¹¹ There is also a need to determine the effectiveness of these controls after they are applied, either permanently or in pilot projects.

This review examines how the Chemical Mass Balance (CMB) receptor model has been used to quantify source contributions from fossil fuel combustion and other sources to ambient concentrations of PM_{2.5} and PM₁₀ for urban and regional scales. Nonfossil fuel sources, such as fugitive dust, cooking, vegetative burn-

* Author to whom correspondence should be addressed.

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ing, and natural or human-caused biogenics must be considered together with fossil-fuel sources in a CMB analysis to obtain closure for $PM_{2.5}$ and PM_{10} mass. The review briefly describes the CMB model, its solution methods, data requirements, and relationship to other receptor and source-oriented models. Recently reported CMB applications that include fossil fuel combustion contributions are tabulated and evaluated. This evaluation leads to generalizations about how the CMB should be used by itself, or preferably in conjunction with other source apportionment methods, to better target air quality improvement strategies for fossil fuel emissions. The evaluation also identifies the types of information that should be published with CMB results that would allow future readers to adequately evaluate the application.

This survey updates and complements, rather than repeats, earlier reviews of databases and CMB applications,^{12–15} limiting itself to studies published since 1995. It assumes the reader is acquainted with the CMB modeling requirements, limitations, validation methods, and results of model testing experiments.^{16,17}

Chemical Mass Balance Receptor Model

The CMB receptor model¹⁸ expresses concentrations of different chemical properties measured at a monitoring site (or "receptor") as a linear sum of products of source profile abundances and source contribution estimates. Source profile abundances are the mass fraction of a chemical or other property in the emissions from each source type that might contribute. Profiles are measured on samples from these sources at times and places believed to represent emissions compositions while receptor measurements are made. Differences in fuels and operating conditions are sought in these tests so that averages and standard deviations of the chemical abundances can be determined. The same properties quantified in source emissions are measured at one or more receptors at sites and over time intervals that correspond with high and low contributions expected from different emitters. They should not interact with each other in nonlinear ways during transport so that the source material adds up to the measured mass at the receptor. All of the source types with a potential for

contributing to the receptor measurements should be identified and had their emissions characterized by methods that simulate their composition in the atmosphere. The relative abundances of these chemical and physical properties need to remain reasonably constant during transport between source and receptor, or important changes need to be simulated. The number of source types used in a CMB source apportionment must be less than or equal to the number of chemical or physical properties measured, and the source profiles must be sufficiently different from one another to be distinguished from each other at the receptor.

Although these requirements seem restrictive, substantial deviations from them can be tolerated in practice, albeit with corresponding increases in the uncertainty of the source contribution estimates. CMB modeling software^{19,20} includes an "effective variance least squares" solution²¹ that propagates measurement precisions for both the ambient data and the source profiles, giving quantitative uncertainty estimates for source contribution estimates. This solution also gives less weight to chemical or physical properties in the ambient concentrations or source profiles that have higher uncertainties or source variability, and larger influence to those source characteristics that are precise and constant. The solution yields large source contribution uncertainties when source profiles are too similar, or "collinear," to be distinguished from each other. Other mathematical tests and performance measures are included to evaluate the goodness of fit, collinearity, and sensitivity to selected model parameters.^{16,17,22}

Another commonly used solution to the CMB equations is the "tracer" solution, in which a particular chemical component is believed to be present only in a single source type and with a fixed ratio to the emitted mass. The carbon-14 isotopic abundance is a tracer compound that distinguishes biogenic (i.e., wood burning, pollen and spores, secondary organic carbon particles formed from monoterpenes emitted by plant life) from fossil fuel contributions to organic and elemental carbon (e.g., Currie et al.,²³ Hawthorne et al.,²⁴ Klinedinst and Currie²⁵). Natural tracers are uncommon, and they cause source type categories to be unnecessarily broad. With carbon-14 by itself, it is not possible to distinguish between contributions from gas, coal, and oil combustion, or between their contributions from different combustion methods. The multivariate effective variance solution provides more robust source contribution estimates because positive biases for some species abundances in a profile are offset by negative

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biases in other abundances. The multivariate solution does not require that a marker species derive from a single source type, only that the ratios among many markers differ among distinct source types.

The CMB has been applied to the apportionment of particulate matter and volatile organic compounds in air,²⁶ contaminants in soils and sediments,^{27–30} wet and dry deposition,^{31,32} and water pollution.^{33–37} Many of these applications involved attribution to fossil fuel sources, especially the raw fuel from oil spills or leaks. The mass balance is the basis for several other receptor models^{38–40} including the enrichment factor, spatial and temporal eigenvector (also termed principal component analysis, factor analysis, or empirical orthogonal function analysis), multiple linear regression, and neural network models.

Receptor models contrast with source-oriented dispersion models^{41,42} that combine source emission rates with meteorological transport and chemical changes to estimate concentrations at a receptor, and with emissions inventories (e.g., U.S. EPA⁴³) that tabulate emissions rates from many sources and are often used to target emissions reductions. Receptor models, source models, and emissions inventories are imperfect representations of reality, and input data are seldom complete. Applying several of these models to the same problem helps to identify and quantify their inaccuracies and to focus further investigation on the areas of greatest discrepancy.⁴⁴

Survey Methodology

Tables of contents and key words from air pollution journals were surveyed for the 1995 through 2001 period

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and copies of articles that might have applied the CMB were obtained and examined. Key word and title searches were made on Cambridge Scientific Abstracts, TheScientificWorld, OCLC, U.S. government documents, and U.S. Environmental Protection Agency (EPA) databases using the key words “CMB”, “chemical mass balance,” “receptor model,” “source apportionment,” “chemical element balance,” and “mass balance.” Scientists and engineers in air quality control agencies that had used receptor models in the past were contacted for information about recent applications.

Many of the publications and reports implicitly used a mass balance in one of the other receptor models identified above. Principal components analysis and factor analysis results were often reported. The major reason given for preferring these methods over the CMB was a lack of source profiles or insufficient chemical measurements to distinguish among sources. Few of these articles produced quantitative source contribution estimates, with most stopping at the source identification step. A more complete summary and evaluation of these other receptor models will be produced in a forthcoming review.

The search found 22 CMB PM_{2.5} and/or PM₁₀ source apportionment studies that are the subject of this review. These apply to data sets acquired from the 1990 through 1998, except for Schauer et al.⁴⁵ who reanalyzed 1982 receptor measurements⁴⁶ using newly measured particulate organic compounds on archived samples.

The following information was extracted from each publication: (1) sampling locations and characteristics (e.g., urban, nonurban, industrial, residential); (2) ambient sampling periods, frequencies, and durations; (3) source categories, source profiles and methods of obtaining profiles; (4) chemical and physical properties quantified at source and receptor; (5) CMB solution and evaluation methods; (6) source contribution estimates; (7) results of reconciliation with source modeling and emissions inventories; (8) novel methods or observations. Authors were consulted when information was missing in a category. The results of the survey were distributed to authors for their review and comment. This information is summarized in Table 1.

Results

Geographic Extent. Even though the CMB is most widely used in the United States, and especially in the western states, it has gained acceptance and application throughout the world. Thirteen of the CMB studies were from the United States, with seven from California's San Francisco Bay area,⁴⁷ San Joaquin Valley,^{48,49} central coast near Santa Barbara,⁵⁰ Los Angeles area,^{45,51} and United States/Mexico border.⁵² The remaining U.S. urban studies were completed in Las Vegas, NV,⁵³ Denver, CO,⁵⁴ and a suburb of Chicago, IL.⁵⁵ Two studies examined source contributions to haze near the

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Table 1. Summary of PM Source Apportionment Studies using CMB and Other Receptor Models

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																								
<p>Reference: San Jose PM₁₀ study.^a</p> <p>When: Two 12-hr daytime (0600 to 1800 PST) and nighttime (1800 to next day 0600 PST) samples were acquired daily from December 1991 to January 1992.</p> <p>Where: San Jose and San Carlos, CA. The San Jose site was in the downtown commercial district. The San Carlos site was 3 km southwest of San Jose in a commercial/residential area.</p> <p>Ambient: Samples were acquired with SFS and analyzed for PM₁₀ mass, 38 elements (Al to Pb by XRF), ions (Cl⁻, NO₃⁻, and SO₄²⁻ by IC; NH₄⁺ by AC; water-soluble Na⁺ and K⁺ by AAS), and carbon (OC and EC by TOR).</p> <p>Source: Samples of paved road dust collected from around the two sites were used to construct geological profiles. These source profiles were reported with associated uncertainties.</p>	<p>Solution: Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <ul style="list-style-type: none"> Transportation (vehicle exhaust)^{c,d} Residential wood combustion^e Marine aerosol^f Secondary aerosol (ammonium sulfate, ammonium nitrate)^g <p>CMB fitting parameters: 26 elements (Al to Pb), ions (Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, K⁺, NH₄⁺), and carbon (OC, EC) from 85 samples were used in CMB attributions to 6 source types.</p> <p>Sensitivity tests: Not reported.</p> <p>Goodness of fit: % mass explained was 100±10% with standard errors of source contribution estimates of 15%-25%.</p> <p>Measurement/modeling uncertainties: Reported.</p> <p>Validation: Comparisons were made between CMB-calculated source contributions to PM₁₀ and estimated emission rates.</p>	<p>Average CMB-calculated source contributions to PM₁₀ (in % mass):</p> <table> <tr> <th>Source Type</th><th>San Jose</th><th>San Carlos</th></tr> <tr> <td>Transportation (vehicle exhaust)</td><td>13%</td><td>13%</td></tr> <tr> <td>Vegetative burning (residential wood combustion [RWC])</td><td>44%</td><td>46%</td></tr> <tr> <td>Geological (paved road dust)</td><td>18</td><td>17%</td></tr> <tr> <td>Marine aerosol</td><td>1.8%</td><td>1.7%</td></tr> <tr> <td>Secondary ammonium sulfate</td><td>3%</td><td>3%</td></tr> <tr> <td>Secondary ammonium nitrate</td><td>18%</td><td>19%</td></tr> <tr> <td>Unexplained</td><td>3%</td><td>2%</td></tr> </table> <p>Average Measured PM₁₀ Mass (μg/m³) 68.4±31.7 64.9±33</p> <p>Number in Average 36 49</p> <p>Contributions to PM₁₀ from nighttime RWC were more than twice the daytime contributions, while vehicle exhaust was 50%-80% higher at night than during the day. Other contributions (e.g., geological, secondary ammonium nitrate) were similar between daytime and nighttime.</p> <p>Large discrepancies were found between emissions inventory and CMB calculations. Inventory estimates vs. CMB calculations of PM₁₀ are 20% vs. 50% for geological, 45% vs. 10% for RWC, and 8% vs. 13% for motor vehicle exhaust, respectively.</p> <p>This study identified needs for: 1) area-specific source profiles for geological, motor vehicle, and residential wood combustion, 2) concurrent measurements of precursor gases for NH₃ and HNO₃, 3) better temporal (shorter duration) and spatial (more sites to represent general population exposure) representation, and 4) reconciliation of source and receptor model results.</p>	Source Type	San Jose	San Carlos	Transportation (vehicle exhaust)	13%	13%	Vegetative burning (residential wood combustion [RWC])	44%	46%	Geological (paved road dust)	18	17%	Marine aerosol	1.8%	1.7%	Secondary ammonium sulfate	3%	3%	Secondary ammonium nitrate	18%	19%	Unexplained	3%	2%
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,i,j}	Source Apportionment Method [‡]	Findings																																																																						
Reference: IMS95 - Integrated Monitoring Study. ^{b,j} When: Daily, 24-hr sampling at the Corcoran core site during fall 1995. Daily, 3-hr, 8 times/day sampling at the Southwest Chowchilla, Fresno, Bakersfield, and Kern Wildlife Refuge core sites during winter 1995-96. Daily, 24-hr sampling at the satellite sites during fall 1995 and winter 1995-96. Where: California's San Joaquin Valley at the Corcoran core site and 22 satellite sites during fall, and at the other 4 core sites and 42 satellite sites during winter. Ambient: PM _{2.5} and PM ₁₀ . Teflon and quartz samples were acquired with SFS and minivol samplers for the winter and fall. Samples were analyzed for mass, <i>b</i> _{abs} (by densitometry), 38 elements (Al to Pb by XRF), ions (Cl ⁻ , NO ₃ ⁻ , and SO ₄ ²⁻ by IC; NH ₄ ⁺ by AC; Na ⁺ and K ⁺ by AAS), and carbon (OC and EC by TOR). Source: No area-specific source profile measurements were taken. Used source profiles collected in central California during 1987-88. ^k	Solution: Effective variance weighted least squares CMB7. ^b Source profiles: Industry (residual oil combustion and oil-field boiler) ^k Transportation (motor vehicle exhaust) ^{c,d,l,m} Vegetative burning (agricultural burning and residential wood combustion) ^k Geological material (agricultural soil, paved and unpaved road dust, ^k and cement dust ⁿ) Marine aerosol ^f Secondary aerosols (ammonium sulfate, ⁿ ammonium nitrate, ⁿ and organics) CMB fitting parameters: 38 elements (Al to Pb), 6 ions, and carbon (OC, EC) from 152 samples were used in CMB attributions to 10 source types. Sensitivity tests: Reported in Magliano et al. ⁱ Goodness of fit: For 224 CMB runs of 3-hr PM _{2.5} samples, R ² = 0.96±0.02, χ ² = 0.67±0.33, and % mass = 90±19%. ⁱ Measurement/modeling uncertainties: Reported in Magliano et al. ⁱ Validation: CMB results were compared with day-specific PM activities and spatially, temporally, and chemically resolved emissions inventories. ^o	Average CMB-calculated source contributions to PM _{2.5} (in % of average mass from Dec. 9-10 and 26-27, 1995, and Jan. 4-6, 1996): <table><tr><th>Source Type</th><th>Southwest Chowchilla</th><th>Fresno</th><th>Bakersfield</th><th>Kern Wildlife Refuge</th></tr><tr><td>Industry (residual oil combustion)</td><td>0.0%</td><td>0.0%</td><td>0.0%</td><td>0.1%</td></tr><tr><td>Transportation (motor vehicle exhaust)</td><td>10.4%</td><td>12.4%</td><td>14.5%</td><td>9.8%</td></tr><tr><td>Vegetative Burning (agricultural burning and residential wood combustion)</td><td>8.4%</td><td>21.0%</td><td>21.7%</td><td>8.9%</td></tr><tr><td>Geological</td><td></td><td></td><td></td><td></td></tr><tr><td>Soil and road dust</td><td>0.4%</td><td>0.3%</td><td>0.7%</td><td>1.0%</td></tr><tr><td>Construction</td><td>0%</td><td>0%</td><td>0%</td><td>0.5%</td></tr><tr><td>Marine aerosol</td><td>0%</td><td>0.2%</td><td>0.4%</td><td>0.1%</td></tr><tr><td>Secondary ammonium sulfate</td><td>7.5%</td><td>3.4%</td><td>5.8%</td><td>7.5%</td></tr><tr><td>Secondary ammonium nitrate</td><td>56.7%</td><td>29.1%</td><td>36.9%</td><td>62.6%</td></tr><tr><td>Excess organics (secondary organics)</td><td>6.3%</td><td>15.3%</td><td>7.2%</td><td>0.9%</td></tr><tr><td>Unexplained</td><td>10.2%</td><td>18.4%</td><td>12.8%</td><td>8.6%</td></tr><tr><td>Average Measured PM_{2.5} Mass (μg/m³)</td><td>30.9</td><td>63.0</td><td>51.7</td><td>28.6</td></tr><tr><td>Number in Average</td><td>56</td><td>56</td><td>56</td><td>56</td></tr></table> Day-specific emissions activities do not correspond to elevated PM ₁₀ concentrations. This suggests that: 1) zones of influence of specific activities are too small to affect ambient concentrations at the site, 2) sampling site is representative of regional or subregional exposure rather than short-term/local influences, and 3) inadequacies of source profiles. Comparisons between emissions inventory and ambient concentrations suggest that the inventory underestimates NMHC and CO and overestimates PM and NO _x . Comparisons between emissions inventory and CMB calculations showed that inventory overestimates geological source emissions while underestimating carbonaceous aerosol.	Source Type	Southwest Chowchilla	Fresno	Bakersfield	Kern Wildlife Refuge	Industry (residual oil combustion)	0.0%	0.0%	0.0%	0.1%	Transportation (motor vehicle exhaust)	10.4%	12.4%	14.5%	9.8%	Vegetative Burning (agricultural burning and residential wood combustion)	8.4%	21.0%	21.7%	8.9%	Geological					Soil and road dust	0.4%	0.3%	0.7%	1.0%	Construction	0%	0%	0%	0.5%	Marine aerosol	0%	0.2%	0.4%	0.1%	Secondary ammonium sulfate	7.5%	3.4%	5.8%	7.5%	Secondary ammonium nitrate	56.7%	29.1%	36.9%	62.6%	Excess organics (secondary organics)	6.3%	15.3%	7.2%	0.9%	Unexplained	10.2%	18.4%	12.8%	8.6%	Average Measured PM _{2.5} Mass (μg/m ³)	30.9	63.0	51.7	28.6	Number in Average	56	56	56	56
Source Type	Southwest Chowchilla	Fresno	Bakersfield	Kern Wildlife Refuge																																																																				
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																																																																								
Reference: Part of IMS95 - Integrated Monitoring Study. ^p When: Daily, 24-hr sampling and daily, 3-hr, 8 times/day sampling from December 1995 to January 1996. Where: California's San Joaquin Valley at two urban sites (Fresno and Bakersfield) and one regional site (Kern Wildlife Refuge). Ambient: 24-hr quartz filter samples acquired with high-volume dichotomous virtual impactor samplers ^q) were analyzed for carbon and organics. ^r 100 organic compounds were quantified, including n-alkanes, n-alkanoic acids, n-alkenoic acids, resin acid, dicarboxylic acids (alkane and aromatic), guaiacols, syringols, and levoglucosin. 3-hr Teflon and quartz samples acquired with SFS were analyzed for mass and chemical composition. ^s Gas-phase hydrocarbon samples acquired by SUVA canisters were analyzed by GC/MS. Source: No area-specific source profile measurements were taken.	<p>Solution: Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <p>Industry (natural gas combustion)^f</p> <p>Transportation (diesel truck, combined organics from Schauer et al.^u with higher fraction EC from Hildemann et al.^w, gasoline vapor [analysis of 1 market weighted average gasoline sample], headspace vapor [calculated based on Conner et al.^x], and natural gas leakage^y)</p> <p>Vegetative burning^z/meat cooking^v</p> <p>Geological (paved road dust)^{p,aa}</p> <p>Secondary aerosol (sulfate, nitrate, organics)</p> <p>CMB fitting parameters: Not reported.</p> <p>Sensitivity tests: Not reported.</p> <p>Goodness of fit: Comparisons of model calculations to measured ambient concentrations for organic compounds are reported.</p> <p>Measurement/modeling uncertainties: Modeling uncertainties were reported.</p> <p>Validation: Both VOCs and particle-phase organic compounds were used to determine source contributions to non-methane organic gases.</p>	<p>Average CMB-calculated annual source contributions to PM_{2.5} (in % mass):</p> <table><thead><tr><th>Source Type</th><th>Fresno (urban)</th><th>Bakersfield (urban)</th><th>Kern Wildlife Refuge (regional)</th></tr></thead><tbody><tr><td>Industry (natural gas combustion)</td><td>1.4% 0**</td><td>0.6% 0.8%</td><td>0** 0*</td></tr><tr><td>Transportation</td><td>12.7% 3.8%</td><td>9.3% 2.0%</td><td>3.6% 0**</td></tr><tr><td>Diesel exhaust</td><td>12.7%</td><td>7.1%</td><td>10.2%</td></tr><tr><td>Gasoline-powered vehicle exhaust</td><td>3.8%</td><td>2.9%</td><td>3.3%</td></tr><tr><td>Vegetative burning/meat cooking</td><td>22.1%</td><td>11.9%</td><td>10.9%</td></tr><tr><td>Hardwood combustion</td><td>41.0%</td><td>2.4%</td><td>8.6%</td></tr><tr><td>Softwood combustion</td><td></td><td></td><td></td></tr><tr><td>Combined wood combustion</td><td></td><td>26.6%</td><td></td></tr><tr><td>Meat cooking</td><td>13.0% 0**</td><td>2.7% 0**</td><td>6.7% 0**</td></tr><tr><td>Vegetative detritus</td><td>0**</td><td>1.6%</td><td>2.2%</td></tr><tr><td>Geological (paved road dust)</td><td>0**</td><td>1.8%</td><td>1.1%</td></tr><tr><td>Sulfate (secondary + background)</td><td>2.5% 21.5%</td><td>4.2% 28.8%</td><td>3.9% 49.6%</td></tr><tr><td>Secondary nitrate</td><td>6.1% 3.1%</td><td>5.1% 9.9%</td><td>6.2% 12.3%</td></tr><tr><td>Secondary ammonium</td><td>9.9% 3.1%</td><td>10.2% 13.7%</td><td>13.8% 14.4%</td></tr><tr><td>Organics (other + secondary)</td><td>7.5%^{††} -27.4%^{‡‡}</td><td>7.8% 14.1%</td><td>13.3% 10.2%</td></tr><tr><td>Unexplained</td><td>0.9% 76.4±4.8^{‡‡}</td><td>5.2% 55.5±3.0</td><td>16.4% 36.3±2.0</td></tr><tr><td>Avg. measured PM_{2.5} mass (µg/m³)</td><td>55.3±3.1</td><td>52.1±2.8</td><td>33.5±1.8</td></tr></tbody></table> <p>** Not statistically different from zero with greater than 95% confidence and therefore removed from CMB model.</p> <p>†† Not statistically different from zero with greater than 95% confidence.</p> <p>‡‡ Ambient particle mass measurement is suspect.</p>	Source Type	Fresno (urban)	Bakersfield (urban)	Kern Wildlife Refuge (regional)	Industry (natural gas combustion)	1.4% 0**	0.6% 0.8%	0** 0*	Transportation	12.7% 3.8%	9.3% 2.0%	3.6% 0**	Diesel exhaust	12.7%	7.1%	10.2%	Gasoline-powered vehicle exhaust	3.8%	2.9%	3.3%	Vegetative burning/meat cooking	22.1%	11.9%	10.9%	Hardwood combustion	41.0%	2.4%	8.6%	Softwood combustion				Combined wood combustion		26.6%		Meat cooking	13.0% 0**	2.7% 0**	6.7% 0**	Vegetative detritus	0**	1.6%	2.2%	Geological (paved road dust)	0**	1.8%	1.1%	Sulfate (secondary + background)	2.5% 21.5%	4.2% 28.8%	3.9% 49.6%	Secondary nitrate	6.1% 3.1%	5.1% 9.9%	6.2% 12.3%	Secondary ammonium	9.9% 3.1%	10.2% 13.7%	13.8% 14.4%	Organics (other + secondary)	7.5% ^{††} -27.4% ^{‡‡}	7.8% 14.1%	13.3% 10.2%	Unexplained	0.9% 76.4±4.8 ^{‡‡}	5.2% 55.5±3.0	16.4% 36.3±2.0	Avg. measured PM _{2.5} mass (µg/m ³)	55.3±3.1	52.1±2.8	33.5±1.8
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{*,†,‡}	Source Apportionment Method [†]	Findings																																																												
		<p>Average CMB-calculated annual source contributions to PM_{2.5} organic carbon (in % mass):</p> <table><tr><th>Source Type</th><th>Bakersfield (urban)</th><th>Fresno (urban)</th><th>Kern Wildlife Refuge (regional)</th></tr><tr><td>Industry (natural gas combustion)</td><td>Dec. 2.2% Jan. 4-6, 1995 0**</td><td>Dec. 2.0% Jan. 4-6, 1995 2.7%</td><td>Dec. 26-28, 1995 0** Jan. 4-6, 1996 0**</td></tr><tr><td>Transportation</td><td></td><td></td><td></td></tr><tr><td>Diesel exhaust</td><td>7.5%</td><td>10.9%</td><td>9.0%</td></tr><tr><td>Gasoline-powered vehicle exhaust</td><td>5.9%</td><td>6.2%</td><td>9.0%</td></tr><tr><td>Vegetative burning/meat cooking</td><td></td><td></td><td></td></tr><tr><td>Hardwood combustion</td><td>21.9%</td><td></td><td>21.2%</td></tr><tr><td>Softwood combustion</td><td>44.0%</td><td></td><td>18.1%</td></tr><tr><td>Combined wood combustion</td><td></td><td>56.4%</td><td>2.2%</td></tr><tr><td>Meat cooking</td><td>14.2%</td><td>5.8%</td><td>14.3%</td></tr><tr><td>Vegetative detritus</td><td>0**</td><td>0**</td><td>0**</td></tr><tr><td>Geological (paved road dust)</td><td>0**</td><td>0.7%</td><td>0**</td></tr><tr><td>Unexplained</td><td>4.3%</td><td>19.9%</td><td>5.7%</td></tr><tr><td>Avg. measured organic mass (µg/m³)[§]</td><td>39.8±2.1</td><td>14.6±0.8</td><td>81.6% 76.1%</td></tr><tr><td></td><td></td><td>13.4±0.7</td><td>4.6±0.3 4.2±0.3</td></tr></table> <p>** Not statistically different from zero with greater than 95% confidence and therefore removed from CMB model.</p> <p>§ Includes the carbon content of secondary organic aerosol. Receptor model results should be less than or equal to the total fine particle organic carbon concentrations.</p> <p>Primary particle emissions from hardwood and softwood combustion, meat cooking, diesel engines, and gasoline-powered motor vehicle exhaust constituted 79% of PM_{2.5} organic aerosol, with small contributions from road dust and natural gas combustion. Combining gas- and particle-phase organic compounds showed that gasoline-powered vehicle exhaust and gasoline vapors are the largest contributors to non-methane organic gases, followed by natural gas leakage.</p>	Source Type	Bakersfield (urban)	Fresno (urban)	Kern Wildlife Refuge (regional)	Industry (natural gas combustion)	Dec. 2.2% Jan. 4-6, 1995 0**	Dec. 2.0% Jan. 4-6, 1995 2.7%	Dec. 26-28, 1995 0** Jan. 4-6, 1996 0**	Transportation				Diesel exhaust	7.5%	10.9%	9.0%	Gasoline-powered vehicle exhaust	5.9%	6.2%	9.0%	Vegetative burning/meat cooking				Hardwood combustion	21.9%		21.2%	Softwood combustion	44.0%		18.1%	Combined wood combustion		56.4%	2.2%	Meat cooking	14.2%	5.8%	14.3%	Vegetative detritus	0**	0**	0**	Geological (paved road dust)	0**	0.7%	0**	Unexplained	4.3%	19.9%	5.7%	Avg. measured organic mass (µg/m ³) [§]	39.8±2.1	14.6±0.8	81.6% 76.1%			13.4±0.7	4.6±0.3 4.2±0.3
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																																																																						
Reference: Santa Barbara PM ₁₀ study. ^{ab} When: 24-hr, every-sixth-day sampling during 1989. Where: Santa Barbara, CA, at 5 urban coastal sites (Santa Barbara, Santa Maria, Santa Ynez, Gaviota, and Watt Island) and 1 offshore background site (Anacapa Island). Ambient: PM ₁₀ Teflon and quartz filter samples were acquired with SFSs. Samples were analyzed for mass, 38 elements (Al to Pb by XRF), ions (Cl ⁻ , NO ₃ ⁻ , and SO ₄ ²⁻ by IC; NH ₄ ⁺ by AC; Na ⁺ and K ⁺ by AAS), and carbon (OC and EC by TOR). Source: 6 local road dust and natural soil samples were acquired, dried, sieved, and subjected to laboratory resuspension ^{ac} to derive site-specific source profiles. Local emissions inventory was used to select profiles for transportation (gas and diesel vehicle exhaust). Marine aerosol was empirically calculated to simulate replacement of sea salt Cl ⁻ by NO ₃ ⁻ at each 25% level. ^{ad} Soil profiles with associated uncertainties are reported.	Solution: Effective variance weighted least squares CMB7. ^b Source profiles: Industry (oil combustion #73501) ^{ae} Transportation (gas and diesel motor vehicle exhaust) ^{af} Geological Limestone ^{ae} Marine aerosol Secondary aerosol (ammonium sulfate and ammonium nitrate) ^g CMB fitting parameters: 17 elements (Al to Pb), ions (NO ₃ ⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , Na ⁺), and carbon (OC, EC) from 330 samples were used in CMB attributions to 6 source types. Sensitivity tests: Reported in Lowenthal et al. ^{ag} Goodness of fit: For 330 CMB runs, R ² = 1.18±0.7, $\chi^2 = 0.97\pm0.02$, and % mass = 93±14%. Measurement/modeling uncertainties: Reported. Validation: NO _x emissions of mobile sources from the emissions inventory was used as a surrogate for HNO ₃ to react with sea salt aerosol to produce HCl and coarse-particle nitrate, and to examine aging of marine aerosol and aerosol acidity.	Average CMB-calculated source contributions to PM ₁₀ (in % mass): <table><tr><th>Source Type</th><th>Santa Barbara</th><th>Santa Maria</th><th>Santa Ynez</th><th>Gaviota</th><th>Watt Road</th><th>Anacapa Island (background)</th></tr><tr><td>Transportation</td><td>42%</td><td>30%</td><td>36%</td><td>25%</td><td>19%</td><td>23%</td></tr><tr><td>Geological</td><td></td><td></td><td></td><td></td><td></td><td></td></tr><tr><td>Road dust/soil</td><td>27%</td><td>29%</td><td>25%</td><td>18%</td><td>9%</td><td>10%</td></tr><tr><td>Limestone</td><td>2%</td><td>0%</td><td>3%</td><td>2%</td><td>0%</td><td>0%</td></tr><tr><td>Marine aerosol</td><td>18%</td><td>23%</td><td>21%</td><td>33%</td><td>55%</td><td>46%</td></tr><tr><td>Secondary ammonium sulfate</td><td>9%</td><td>12%</td><td>12%</td><td>15%</td><td>11%</td><td>16%</td></tr><tr><td>Secondary ammonium nitrate</td><td>3%</td><td>6%</td><td>3%</td><td>3%</td><td>6%</td><td>5%</td></tr><tr><td>Average measured PM₁₀ mass (μg/m³)</td><td>34±2</td><td>27±2</td><td>19.0±1.3</td><td>20.5±1.4</td><td>20.6±1.3</td><td>26±2</td></tr><tr><td>Number in average</td><td>59</td><td>60</td><td>60</td><td>58</td><td>58</td><td>35</td></tr></table> Fresh marine aerosols were acidic, accompanied by reduced concentrations of SO ₄ ²⁻ , NO ₃ ⁻ , and lower-level urban emissions of motor vehicles. Aged marine aerosols were characterized by a deficit of Cl ⁻ with respect to sea salt, elevated (NH ₄) ₂ SO ₄ , NH ₄ NO ₃ , and higher levels of urban emissions of vehicle exhaust.	Source Type	Santa Barbara	Santa Maria	Santa Ynez	Gaviota	Watt Road	Anacapa Island (background)	Transportation	42%	30%	36%	25%	19%	23%	Geological							Road dust/soil	27%	29%	25%	18%	9%	10%	Limestone	2%	0%	3%	2%	0%	0%	Marine aerosol	18%	23%	21%	33%	55%	46%	Secondary ammonium sulfate	9%	12%	12%	15%	11%	16%	Secondary ammonium nitrate	3%	6%	3%	3%	6%	5%	Average measured PM ₁₀ mass (μg/m ³)	34±2	27±2	19.0±1.3	20.5±1.4	20.6±1.3	26±2	Number in average	59	60	60	58	58	35
Source Type	Santa Barbara	Santa Maria	Santa Ynez	Gaviota	Watt Road	Anacapa Island (background)																																																																		
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Marine aerosol	18%	23%	21%	33%	55%	46%																																																																		
Secondary ammonium sulfate	9%	12%	12%	15%	11%	16%																																																																		
Secondary ammonium nitrate	3%	6%	3%	3%	6%	5%																																																																		
Average measured PM ₁₀ mass (μg/m ³)	34±2	27±2	19.0±1.3	20.5±1.4	20.6±1.3	26±2																																																																		
Number in average	59	60	60	58	58	35																																																																		

Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [†]	Findings																																																																			
<p>Reference: South Coast Air Quality Management District PTEP (PM₁₀ Technology Enhancement Program).^{ai}</p> <p>When: 24-hr sampling on a schedule of daily (second half of 1995), every third day (second quarter of 1995), and every sixth day (1st quarter of 1995) at all sites except San Nicolas Island, which sampled every sixth day all year.</p> <p>Where: At 6 locations (downtown Los Angeles, Anaheim, Diamond Bar, Rubidoux, Fontana, and upwind San Nicolas Island background site) in southern California.</p> <p>Ambient: 24-hr Teflon and quartz filter samples were acquired with modified SCAQS samplers.</p> <p>Samples were analyzed for mass, 36 elements (by XRF), ions (Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, and K⁺ by IC), and carbon (OC and EC by TOR).</p> <p>Source: No area-specific source profile measurements were taken during the study. Used SCAQS source profiles acquired in the early 1980s.^{ai}</p>	<p>Solution: Principal component analysis and correlation matrix were used to select major source types for effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <p>Industry (residual oil combustion)</p> <p>Transportation (motor vehicle exhaust)</p> <p>Geological (paved and unpaved road dust, limestone)</p> <p>Marine aerosol</p> <p>Secondary aerosol (ammonium sulfate, ammonium nitrate, organics)</p> <p>CMB fitting parameters: 20 elements, 5 ions (NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺), and carbon (OC, EC) from 24-hr samples were used in CMB attributions to as many as 8 source types.</p> <p>Sensitivity tests: Not reported.</p> <p>Goodness of fit: Not reported.</p> <p>Measurement/modeling uncertainties: Not reported.</p> <p>Validation: Urban airshed models (UAM/LC and UAM-AERO) were used to simulate PM₁₀ episodes and to project future-year air quality. Future air quality was estimated using UAM/LC and CMB with different emission control scenarios.</p>	<p>Average CMB-calculated source contributions to annual PM₁₀ in southern California (in % mass):</p> <table><thead><tr><th rowspan="2">Source Type</th><th colspan="3">Downtown Los Angeles</th><th colspan="2">Diamond Bar</th><th>Fontana</th><th>Rubidoux</th></tr><tr><th>Los Angeles</th><th>Anaheim</th><th>Bar</th><th>Fontana</th><th>Rubidoux</th></tr></thead><tbody><tr><td>Industry (residual oil combustion)</td><td>3.6%</td><td>5.3%</td><td>3.8%</td><td>2.3%</td><td>1.7%</td></tr><tr><td>Transportation (motor vehicle exhaust)</td><td>13.4%</td><td>11.1%</td><td>10.9%</td><td>7.5%</td><td>6.9%</td></tr><tr><td>Geological material</td><td>36%</td><td>30.2%</td><td>30.5%</td><td>49.4%</td><td>39.3%</td></tr><tr><td>Limestone</td><td>0%</td><td>0%</td><td>0%</td><td>0%</td><td>8.2%</td></tr><tr><td>Marine aerosol</td><td>4.2%</td><td>5.8%</td><td>4.5%</td><td>2.5%</td><td>2.3%</td></tr><tr><td>Secondary ammonium sulfate</td><td>10.6%</td><td>11.2%</td><td>10.5%</td><td>6.3%</td><td>6.6%</td></tr><tr><td>Secondary ammonium nitrate</td><td>24.1%</td><td>29.3%</td><td>31.8%</td><td>25.6%</td><td>28.2%</td></tr><tr><td>Secondary organics</td><td>8.1%</td><td>7.0%</td><td>8.0%</td><td>6.4%</td><td>6.8%</td></tr><tr><td>Average Measured PM₁₀ Mass (μg/m³)</td><td>48.1±3.1</td><td>42.3±4.2</td><td>46.8±4.7</td><td>64.8±4.6</td><td>75.7±6.2</td></tr></tbody></table> <p>Large spatial variations in PM₁₀ source contributions were found for geological material and vehicle exhaust. Fossil fuel contributions from residual oil combustion are small (in the range of 2%-5%).</p>	Source Type	Downtown Los Angeles			Diamond Bar		Fontana	Rubidoux	Los Angeles	Anaheim	Bar	Fontana	Rubidoux	Industry (residual oil combustion)	3.6%	5.3%	3.8%	2.3%	1.7%	Transportation (motor vehicle exhaust)	13.4%	11.1%	10.9%	7.5%	6.9%	Geological material	36%	30.2%	30.5%	49.4%	39.3%	Limestone	0%	0%	0%	0%	8.2%	Marine aerosol	4.2%	5.8%	4.5%	2.5%	2.3%	Secondary ammonium sulfate	10.6%	11.2%	10.5%	6.3%	6.6%	Secondary ammonium nitrate	24.1%	29.3%	31.8%	25.6%	28.2%	Secondary organics	8.1%	7.0%	8.0%	6.4%	6.8%	Average Measured PM ₁₀ Mass (μg/m ³)	48.1±3.1	42.3±4.2	46.8±4.7	64.8±4.6	75.7±6.2
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{*,†,‡}	Source Apportionment Method [†]	Findings																																																																																										
Reference: Source apportionment of PM ₂ with organic markers. [†] When: 24-hr, every-sixth-day sampling during 1982. Where: At 4 locations (Pasadena, downtown Los Angeles, West Los Angeles, and Rubidoux) in southern California. Ambient: Teflon, quartz (on 2 parallel channels), and Nuclepore filter samples were acquired with Caltech PM ₂ samplers. ^{aj} Samples were analyzed for mass, 34 elements (by XRF), ions (NO ₃ and SO ₄ by IC), and carbon (OC and EC by TOR). The second channel's quartz filter is composited by each month for quantification of individual organic compounds by HRGC and GC/MS. Source: No area-specific source profile measurements were taken. Source samples of 15 source types were acquired from previous studies. ^{†,w,z,ak}	Solution: Effective variance weighted least squares CMB7. ^b Source profiles: ^{†,w,z,ak} Industry (natural gas combustion, surface coating, roofing tar pots, misc. industrial point sources) Transportation (non-catalyst gasoline vehicles, catalyst-equipped gasoline vehicles, diesel vehicles, tire wear debris, brake lining wear debris) Vegetative burning (fireplaces, forest fires, meat cooking) Geological (paved road dust) Cigarettes Secondary aerosol (sulfate, nitrate, organics [other + secondary]) CMB fitting parameters: 12 source profiles and 45 organic compounds plus EC, Al, and Si were used as model input. Sensitivity tests: Not reported. Goodness of fit: Comparison of model calculations to measured ambient concentrations for organic compounds are reported. Validation: Ratios of annual average ambient concentrations to emission rates in the inventory for each organic component were used to test for chemical stability and to select a subset of organic markers for CMB calculations (average ± 2 standard deviations of the ratios of stable species, EC, Al, and Si were used to set up the interval). Final selection of organic species for model input was determined by stability test results and data from other studies.	Average CMB-calculated annual source contributions to PM ₂ (in % mass): <table><thead><tr><th>Source Type</th><th>Pasadena</th><th>Downtown Los Angeles</th><th>West Los Angeles</th><th>Rubidoux</th></tr></thead><tbody><tr><td>Industry (natural gas combustion aerosol)</td><td>0.2%</td><td>0.1%</td><td>0.1%</td><td>0.1%</td></tr><tr><td>Transportation</td><td></td><td></td><td></td><td></td></tr><tr><td> Diesel exhaust</td><td>18.7%</td><td>35.7%</td><td>17.8%</td><td>12.7%</td></tr><tr><td> Catalyst and non-catalyst gasoline-powered vehicle exhaust</td><td>5.8%</td><td>6.5%</td><td>5.9%</td><td>0.8%</td></tr><tr><td> Tire wear debris</td><td>1.0%</td><td>0.7%</td><td>1.0%</td><td>0**</td></tr><tr><td>Vegetative burning/meat cooking</td><td></td><td></td><td></td><td></td></tr><tr><td> Wood smoke</td><td>9.6%</td><td>5.7%</td><td>10.8%</td><td>1.3%</td></tr><tr><td> Meat charbroiling and frying</td><td>8.5%</td><td>5.4%</td><td>8.3%</td><td>4.6%</td></tr><tr><td> Vegetative detritus</td><td>1.2%</td><td>0.7%</td><td>1.6%</td><td>0.4%</td></tr><tr><td> Geological (paved road dust)</td><td>12.3%</td><td>11.1%</td><td>12.2%</td><td>13.1%</td></tr><tr><td> Sulfate (secondary + background)</td><td>20.9%</td><td>20.3%</td><td>24.1%</td><td>13.8%</td></tr><tr><td> Secondary nitrate</td><td>7.4%</td><td>9.2%</td><td>7.8%</td><td>24.7%</td></tr><tr><td> Secondary ammonium</td><td>9.2%</td><td>9.2%</td><td>9.4%</td><td>12.1%</td></tr><tr><td> Organics (other + secondary)</td><td>5.2%</td><td>3.6%</td><td>4.2%^{††}</td><td>4.6%</td></tr><tr><td> Other sources (cigarette smoke)</td><td>0.6%</td><td>0.8%</td><td>0.8%</td><td>0.5%</td></tr><tr><td>Unexplained</td><td>-0.4%</td><td>-9.2%</td><td>-3.3%</td><td>-11.4%</td></tr><tr><td>Average measured mass (µg/m³)</td><td>28.2±1.9[^]</td><td>32.5±2.8[^]</td><td>24.5±2.0[^]</td><td>42.1±3.3</td></tr></tbody></table> ** Not statistically different from zero with greater than 95% confidence and therefore removed from CMB model. †† Includes secondary organic aerosol; receptor model results should be less than or equal to measured total fine particle organic concentration. ^ Measured and apportioned values are not significantly different using a 95% confidence interval.	Source Type	Pasadena	Downtown Los Angeles	West Los Angeles	Rubidoux	Industry (natural gas combustion aerosol)	0.2%	0.1%	0.1%	0.1%	Transportation					Diesel exhaust	18.7%	35.7%	17.8%	12.7%	Catalyst and non-catalyst gasoline-powered vehicle exhaust	5.8%	6.5%	5.9%	0.8%	Tire wear debris	1.0%	0.7%	1.0%	0**	Vegetative burning/meat cooking					Wood smoke	9.6%	5.7%	10.8%	1.3%	Meat charbroiling and frying	8.5%	5.4%	8.3%	4.6%	Vegetative detritus	1.2%	0.7%	1.6%	0.4%	Geological (paved road dust)	12.3%	11.1%	12.2%	13.1%	Sulfate (secondary + background)	20.9%	20.3%	24.1%	13.8%	Secondary nitrate	7.4%	9.2%	7.8%	24.7%	Secondary ammonium	9.2%	9.2%	9.4%	12.1%	Organics (other + secondary)	5.2%	3.6%	4.2% ^{††}	4.6%	Other sources (cigarette smoke)	0.6%	0.8%	0.8%	0.5%	Unexplained	-0.4%	-9.2%	-3.3%	-11.4%	Average measured mass (µg/m ³)	28.2±1.9 [^]	32.5±2.8 [^]	24.5±2.0 [^]	42.1±3.3
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<p>Reference: Imperial Valley/Mexicali PM₁₀ study.^{al}</p> <p>When: 24-hr, every-sixth-day sampling from March 1992 to August 1993. Daily, 6-hr (starting at 0000 PST), 4 times/day sampling during weeklong spring, summer, and winter intensive sampling periods.</p> <p>Where: Imperial/Mexicali Valley (189 km inland from the Pacific Coast) at two border cities: Calexico, California [U.S.], and Mexicali, Baja California [Mexico] (~10 km south of Calexico).</p> <p>Weeklong intensive sampling was also conducted at 20 satellite sites.</p> <p>Ambient: Teflon and quartz filter samples were acquired with SFS and minivol samplers. Samples were analyzed for mass, b_{abs} (by densitometry), 40 elements (Na to U by XRF), ions (Cl⁻, NO₃⁻, and SO₄²⁻ by IC; NH₄⁺ by AC; and water-soluble Na⁺ and K⁺ by AAS), and carbon (OC, EC by TOR).</p> <p>Source: 7 industrial (5 manure-fired power plant, 2 glass manufacturing), 11 roadside motor vehicle exhaust along the U.S./Mexico border, 1 vegetative burning, 2 charcoal meat cooking, and 5 fugitive dust source samples were collected.^{am} Source profiles applied are reported with associated uncertainties in Watson and Chow.^{am}</p>	<p>Solution: Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <ul style="list-style-type: none"> Industry (manure-fired power plant, glass manufacturing plant) Transportation Vegetative burning (asparagus burn, residential wood combustion with fireplace^e and woodstove^a, and charcoal cooking) Geological material (soil, road dust, and limestone) Secondary aerosol (ammonium sulfate and ammonium nitrate)^g <p>CMB fitting parameters: 18 elements (Na to Pb), ions (NO₃⁻, SO₄²⁻, Na⁺, K⁺, NH₄⁺), and carbon (OC and EC) from 254 24-hr samples and 192 6-hr samples were used in CMB attributions to 6 source types.</p> <p>Sensitivity tests: Reported in Chow and Watson.^{am}</p> <p>Goodness of fit: Reported in Chow and Watson.^{am}</p> <p>Measurement/modeling uncertainties: Reported.</p> <p>Validation: Wind trajectory was used to calculate cross-border flux of pollutant transport.</p>	<p>Average CMB-calculated source contributions to PM₁₀ (in % mass) between Sept. 3, 1992, and Aug. 29, 1993:</p> <table> <tr> <th>Source Type</th><th>Calexico, CA</th><th>Mexicali, Baja California, Mexico</th></tr> <tr> <td>Industry</td><td></td><td></td></tr> <tr> <td>Manure-fueled power plant</td><td>0.03%</td><td>0%</td></tr> <tr> <td>Glass manufacturing plant</td><td>0.07%</td><td>0.27%</td></tr> <tr> <td>Transportation (motor vehicle exhaust)</td><td>16.0%</td><td>12.9%</td></tr> <tr> <td>Vegetative/charbroil burning</td><td>4.3%</td><td>7.6%</td></tr> <tr> <td>Geological (composite soil and paved road dust)</td><td>72.9%</td><td>70.4%</td></tr> <tr> <td>Marine aerosol</td><td>2.9%</td><td>1.7%</td></tr> <tr> <td>Secondary ammonium sulfate</td><td>3.3%</td><td>1.5%</td></tr> <tr> <td>Secondary ammonium nitrate</td><td>2.4%</td><td>1.5%</td></tr> <tr> <td>Unexplained</td><td>-1.9%</td><td>4.2%</td></tr> <tr> <td>Average Measured PM₁₀ Mass (μg/m³)</td><td>61.9±4.5</td><td>129.9±8.9</td></tr> <tr> <td>Number in Average</td><td>55</td><td>48</td></tr> </table> <p>Source contribution estimates (SCE) were used to understand diurnal variations of 6-hr (4 times/day) samples in each season. Three- to fourfold differences in SCE are found, with low contributions in the afternoon (1200-1800 PST) and high contributions at night (1800-2400 PST) during winter and spring for motor vehicle exhaust and geological material. During spring, the highest source contributions occur during the morning period (0600-1200 PST), and the lowest during the early morning period (0000-0600 PST).</p>	Source Type	Calexico, CA	Mexicali, Baja California, Mexico	Industry			Manure-fueled power plant	0.03%	0%	Glass manufacturing plant	0.07%	0.27%	Transportation (motor vehicle exhaust)	16.0%	12.9%	Vegetative/charbroil burning	4.3%	7.6%	Geological (composite soil and paved road dust)	72.9%	70.4%	Marine aerosol	2.9%	1.7%	Secondary ammonium sulfate	3.3%	1.5%	Secondary ammonium nitrate	2.4%	1.5%	Unexplained	-1.9%	4.2%	Average Measured PM ₁₀ Mass (μg/m ³)	61.9±4.5	129.9±8.9	Number in Average	55	48
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Secondary ammonium nitrate	2.4%	1.5%																																							
Unexplained	-1.9%	4.2%																																							
Average Measured PM ₁₀ Mass (μg/m ³)	61.9±4.5	129.9±8.9																																							
Number in Average	55	48																																							

Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																																																																								
Reference: Las Vegas PM ₁₀ Study. ^{ao} When: 24-hr, every-sixth-day sampling from January 1995 to January 1996. Daily sampling during the winter holiday period (Dec. 23, 1995, to Jan. 4, 1996), and during five seasonal weeklong intensive sampling periods (2 spring, 1 summer, 1 fall, and 1 winter). Where: Las Vegas Valley, NV, at two base sites. The Bemis site was near construction, and the East Charleston site was near neighborhoods and roads. Intensive sampling was conducted at 29 satellite sites. Ambient: Teflon and quartz filter samples were acquired with: 1) SFS and analyzed for mass, b _{abs} (by densitometry), 38 elements (Al to Pb by XRF), ions (Cl ⁻ , NO ₃ ⁻ , and SO ₄ ²⁻ by IC; NH ₄ ⁺ by AC; and water-soluble K ⁺ by AAS), and carbon (OC and EC by TOR), and 2) Minivol and analyzed for mass, b _{abs} , and crustal elements. Source: Source profiles were acquired for transportation (20 roadside samples of a mixture of vehicles), vegetative burning (22 samples from 4 fireplaces and 1 woodstove), and geological (22 samples from paved and unpaved roads, paved parking lot, construction site, and desert soil). Source profiles applied were reported with associated uncertainties in Chow and Watson. ^{ap}	Solution: Effective variance weighted least squares CMB7 receptor modeling ^b and ISCST-3 dispersion modeling for source/receptor model reconciliation. Source profiles: Transportation (roadside gasoline and diesel vehicle exhaust) Vegetative burning (residential wood combustion from fireplaces and woodstoves) Geological (paved and unpaved road dust, construction dust, and desert soil) Secondary aerosol (ammonium sulfate, ammonium nitrate) ^g CMB fitting parameters: 18 elements (Na to Pb), ions (NO ₃ ⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , K ⁺), and carbon (OC, EC) from 98 samples from each of the 2 base sites were used in CMB attribution to 5 source types. Sensitivity tests: Reported in Chow and Watson. ^{ap} Goodness of fit: Reported in Chow and Watson. ^{ap} Measurement/modeling uncertainties: Reported for CMB, but not for ISCST-3. Validation: ISCST-3 model calculations (using 1995 emissions inventory of 9 source types with surface meteorology data from 16 locations in the Las Vegas Valley) were compared with CMB results and emissions inventory estimates.	Comparison of PM ₁₀ source and receptor modeling results (in % mass): <table><thead><tr><th>Source Type</th><th>Bemis site CMB ISCST-3</th><th>East Charleston site CMB ISCST-3</th><th>1995 Emissions Inventory</th></tr></thead><tbody><tr><td>Transportation (gasoline and diesel exhaust)</td><td>2.7%</td><td>1%</td><td>7%</td></tr><tr><td>Residential wood combustion</td><td>0.2%</td><td>0.3%</td><td>2%</td></tr><tr><td>Geological material</td><td>93.3%</td><td>99%</td><td>96.4%</td></tr><tr><td>Soil and paved road dust</td><td>NA</td><td>51%</td><td>23%</td></tr><tr><td>Construction activity</td><td>NA</td><td>10%</td><td>7%</td></tr><tr><td>Construction trackout</td><td>NA</td><td>8%</td><td>6%</td></tr><tr><td>Construction wind erosion</td><td>NA</td><td>7%</td><td>43%</td></tr><tr><td>Paved roads</td><td>NA</td><td>7%</td><td>7.04%</td></tr><tr><td>Unpaved roads</td><td>NA</td><td>15%</td><td>7%</td></tr><tr><td>Disturbed land</td><td>NA</td><td>NA</td><td>NA</td></tr><tr><td>Rural wind erosion</td><td>NA</td><td>NA</td><td>36%</td></tr><tr><td>Stationary sources</td><td>NA</td><td>0.5%</td><td>2.13%</td></tr><tr><td>Permitted off-road racing</td><td>NA</td><td>NA</td><td>0.19%</td></tr><tr><td>Secondary ammonium sulfate</td><td>1.9%</td><td>NA</td><td>NA</td></tr><tr><td>Secondary ammonium nitrate</td><td>1.9%</td><td>NA</td><td>NA</td></tr><tr><td>Average measured PM₁₀ Mass</td><td>34.6±2.1 μg/m³</td><td>183.4 μg/m³</td><td>44.7±2.6 μg/m³</td></tr><tr><td>Number in average:</td><td>98</td><td>98</td><td>98</td></tr></tbody></table> 1995 emissions inventory agrees qualitatively with CMB receptor model in attributing >80% of PM ₁₀ to fugitive dust sources in the Las Vegas Valley, but the ISCST-3 model results in 2 to 4 times higher absolute PM ₁₀ concentrations. This is probably due to the methodology used for windblown dust emission calculations in the GIS-based emissions inventory. 1995 emissions inventory and ISCST-3 model underestimated contributions from transportation to PM ₁₀ .	Source Type	Bemis site CMB ISCST-3	East Charleston site CMB ISCST-3	1995 Emissions Inventory	Transportation (gasoline and diesel exhaust)	2.7%	1%	7%	Residential wood combustion	0.2%	0.3%	2%	Geological material	93.3%	99%	96.4%	Soil and paved road dust	NA	51%	23%	Construction activity	NA	10%	7%	Construction trackout	NA	8%	6%	Construction wind erosion	NA	7%	43%	Paved roads	NA	7%	7.04%	Unpaved roads	NA	15%	7%	Disturbed land	NA	NA	NA	Rural wind erosion	NA	NA	36%	Stationary sources	NA	0.5%	2.13%	Permitted off-road racing	NA	NA	0.19%	Secondary ammonium sulfate	1.9%	NA	NA	Secondary ammonium nitrate	1.9%	NA	NA	Average measured PM ₁₀ Mass	34.6±2.1 μg/m ³	183.4 μg/m ³	44.7±2.6 μg/m ³	Number in average:	98	98	98
Source Type	Bemis site CMB ISCST-3	East Charleston site CMB ISCST-3	1995 Emissions Inventory																																																																							
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																																																																																																																																																																											
<p>Reference: NFRAQS – Northern Front Range Air Quality Study.^{aq}</p> <p>When: 6-hr (4 times/day), 12-hr (2 times/day), and 24-hr daily sampling during winter and summer 1996 and winter 1997.</p> <p>Where: At 9 locations in Colorado, including 3 urban city center sites (Fort Collins, Welby [Denver], CAMP [Denver]), 2 rural river valley sites (Evans, Masters), 1 rural down-valley site (Brighton), 1 regional transport site (Longmont), 1 upwind commercial/residential site (Highlands), and 1 rural site (Chatfield).</p> <p>Ambient: PM_{2.5} and gaseous (NH₃, HNO₃) samples were acquired with SFS, SGS, and minivol samplers. Samples were analyzed for mass, 38 elements (Al to Pb by XRF), ions (Cl⁻, NO₃⁻, and SO₄²⁻ by IC; NH₄⁺ by AC; Na⁺ and K⁺ by AAS), carbon (OC and EC by TOR), and C-14 (by accelerator mass spectrometry^{at}).</p> <p>Source: Source profiles for transportation (spark-ignition and diesel exhaust), residential wood combustion, meat cooking, geological material, and C-14 were acquired. Source profiles for the study are reported with associated uncertainties in Zielinska et al.^{as}</p>	<p>Solution: Effective variance weighted least squares CMB8.^{at}</p> <p>Source profiles:</p> <p>Industry (coal-fired power plant)^e</p> <p>Transportation (light-duty gasoline vehicles [cold start, hot stabilized, high emitters], diesel exhaust)</p> <p>Vegetative burning (meat cooking, softwood and hardwood combustion)</p> <p>Geological (paved road dust, soil)</p> <p>Secondary aerosol (ammonium sulfate, ammonium nitrate)^g</p> <p>CMB fitting parameters: 25 elements (Al to Pb), ions (Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺, K⁺), carbon (OC, EC), and 85 organic species from 150 24-hr samples were used in conventional CMB run, and from 132 6- and 12-hr samples in extended species CMB runs.^{au}</p> <p>Sensitivity tests: Reported in Fujita et al.^{au}</p> <p>Goodness of fits: Typical R² > 0.9, $\chi^2 < 0.5$, and % mass = 100±20%.</p> <p>Measurement/modeling uncertainties: Reported.</p> <p>Validation: Conventional CMB and speciated CMB (with detailed organic speciation) calculations were made to estimate source contributions to PM_{2.5} mass, carbon, and gases (SO₂, NO_x).</p>	<p>Average CMB-calculated source contributions to PM_{2.5}, carbon, SO₂, and NO_x (in % mass) at the Welby site:</p> <table><thead><tr><th rowspan="2">Source Type</th><th colspan="2">PM_{2.5}</th><th colspan="2">Total Carbon</th><th colspan="2">SO₂</th><th colspan="2">NO_x</th><th rowspan="2">1995 Emissions Inventory</th></tr><tr><th>Conventional CMB</th><th>Extended Species CMB</th><th>Conventional CMB</th><th>Extended Species CMB</th><th>Conventional CMB</th><th>Extended Species CMB</th><th>Conventional CMB</th><th>Extended Species CMB</th></tr></thead><tbody><tr><td>Industry (coal-fired power plant)</td><td>0.8%</td><td>2.3%</td><td>0.1%</td><td>0.2%</td><td>51.4%</td><td>7.2%</td><td>2.6%</td><td></td></tr><tr><td>Transportation</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></tr><tr><td> Light-duty gasoline vehicle (LDGV)</td><td>31.3%</td><td></td><td>60.8%</td><td></td><td></td><td></td><td>5.8%</td><td></td></tr><tr><td> LDGV cold start</td><td></td><td>11.9%</td><td></td><td>24.0%</td><td></td><td>11.2%</td><td></td><td>15.3%</td></tr><tr><td> LDGV hot stabilized</td><td></td><td>3.2%</td><td></td><td>6.6%</td><td></td><td>7.8%</td><td></td><td>11.7%</td></tr><tr><td> LDGV high particle emitter</td><td></td><td>12.7%</td><td></td><td>28.3%</td><td></td><td>2.9%</td><td></td><td>6.6%</td></tr><tr><td> Diesel exhaust</td><td>9.0%</td><td>10.0%</td><td>19.8%</td><td>22.0%</td><td>6.1%</td><td>17.0%</td><td>17.8%</td><td></td></tr><tr><td>Vegetative burning/meat cooking</td><td>6.0%</td><td></td><td>15.0%</td><td></td><td></td><td></td><td></td><td></td></tr><tr><td> Meat cooking</td><td></td><td>3.7%</td><td></td><td>6.3%</td><td></td><td>0.0%</td><td></td><td>0.0%</td></tr><tr><td> Wood combustion (softwood)</td><td></td><td>2.1%</td><td></td><td>3.8%</td><td></td><td>0.0%</td><td></td><td>0.0%</td></tr><tr><td> Wood combustion (hardwood)</td><td></td><td>2.7%</td><td></td><td>4.2%</td><td></td><td>0.1%</td><td></td><td>0.0%</td></tr><tr><td>Geological material (paved road dust)</td><td>16.7%</td><td></td><td>4.3%</td><td></td><td>4.2%</td><td></td><td></td><td>43.24%</td></tr><tr><td>Secondary ammonium sulfate</td><td>9.6%</td><td></td><td>0.0%</td><td></td><td></td><td></td><td></td><td></td></tr><tr><td>Secondary ammonium nitrate</td><td>23.2%</td><td></td><td>0.0%</td><td></td><td></td><td></td><td></td><td></td></tr><tr><td>Unexplained</td><td>3.4%</td><td>1.7%</td><td>0.0%</td><td>0.4%</td><td>20.4%</td><td>42.1%</td><td></td><td></td></tr><tr><td>Average measured mass</td><td>14.6±1.0 µg/m³</td><td>16.3±1.0 µg/m³</td><td>6.2±0.4 µg/m³</td><td>6.5±0.6 µg/m³</td><td>13.3±5.3 µg/m³</td><td>214.7±28.3 µg/m³</td><td>27.5 tons/day</td><td></td></tr><tr><td>Number in Average</td><td>16</td><td>15</td><td>16</td><td>17</td><td>18</td><td>17</td><td></td><td></td></tr></tbody></table> <p>** Assuming half softwood and half hardwood.</p> <p>CMB calculations show that coal-fired power plants contribute 2% of primary PM_{2.5}, ~7%-9% of NO_x, ~10%-15% of ground-level NO_x and NO₃⁻, and 41%-50% of SO₂ in Denver. Motor vehicle exhaust contributes 55% of PM_{2.5}, 51%-73% of NO_x, and 22%-28% of SO₂.</p> <p>C-14 shows that 80% of geological material and 100% of meat cooking and woodburning emissions consist of contemporary carbon, while vehicle exhaust consists of 100% fossil carbon.</p> <p>Comparisons between emissions inventory and CMB calculations of carbon are 20% vs. 68% for gasoline vehicle exhaust contributions, and 80% vs. 32% for diesel exhaust contributions. LDGV cold starts (especially for older vehicles) and LDGV high emitters account for most of the gasoline vehicle exhaust emissions.</p>	Source Type	PM _{2.5}		Total Carbon		SO ₂		NO _x		1995 Emissions Inventory	Conventional CMB	Extended Species CMB	Conventional CMB	Extended Species CMB	Conventional CMB	Extended Species CMB	Conventional CMB	Extended Species CMB	Industry (coal-fired power plant)	0.8%	2.3%	0.1%	0.2%	51.4%	7.2%	2.6%		Transportation									Light-duty gasoline vehicle (LDGV)	31.3%		60.8%				5.8%		LDGV cold start		11.9%		24.0%		11.2%		15.3%	LDGV hot stabilized		3.2%		6.6%		7.8%		11.7%	LDGV high particle emitter		12.7%		28.3%		2.9%		6.6%	Diesel exhaust	9.0%	10.0%	19.8%	22.0%	6.1%	17.0%	17.8%		Vegetative burning/meat cooking	6.0%		15.0%						Meat cooking		3.7%		6.3%		0.0%		0.0%	Wood combustion (softwood)		2.1%		3.8%		0.0%		0.0%	Wood combustion (hardwood)		2.7%		4.2%		0.1%		0.0%	Geological material (paved road dust)	16.7%		4.3%		4.2%			43.24%	Secondary ammonium sulfate	9.6%		0.0%						Secondary ammonium nitrate	23.2%		0.0%						Unexplained	3.4%	1.7%	0.0%	0.4%	20.4%	42.1%			Average measured mass	14.6±1.0 µg/m ³	16.3±1.0 µg/m ³	6.2±0.4 µg/m ³	6.5±0.6 µg/m ³	13.3±5.3 µg/m ³	214.7±28.3 µg/m ³	27.5 tons/day		Number in Average	16	15	16	17	18	17		
Source Type	PM _{2.5}			Total Carbon		SO ₂		NO _x		1995 Emissions Inventory																																																																																																																																																																			
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,1,†}	Source Apportionment Method [†]	Findings																																							
<p>Reference: Robbins Particulate Study.^{av}</p> <p>When: 24-hr, every-sixth-day sampling from 1995 to 1997.</p> <p>Where: At 4 locations (Alsip, Berman, Meadow Lane, and Eisenhower) near the Robbins Resource Recovery Facility (RRRF) in the metropolitan Chicago, Illinois, area.</p> <p>Ambient: PM_{2.5} and PM_{coarse} Teflon filter samples were acquired with dichotomous samplers. 24-hr PM₁₀ quartz filter samples were acquired with hi-vol-SSI samplers. Samples were analyzed for mass, 40 elements (Na to U by XRF), ions (Cl⁻, NO₃⁻, and SO₄²⁻ by IC; NH₄⁺ by AC; Na⁺ and K⁺ by AAS), and carbon (OC and EC by TOR).</p> <p>Source: Source profiles were acquired for Waste-To-Energy (WTE) fly ash, dust from the refuse-derived fuel (RDF) area, oil refinery catalyst, and road salt. Source profiles for the study with associated uncertainties are reported in Appendix B of Watson et al.^{av}</p>	<p>Solution: Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <ul style="list-style-type: none"> Industry (power plant,^{aw} WTE incinerator, steel plant^{ax,ay}) Transportation (vehicle exhaust)^{az} Vegetative burning (residential wood combustion)^{ba} Geological (local road dust) Road salt Secondary aerosol (ammonium sulfate, ammonium nitrate)^{be} Pure species profiles (100% of each element) of Zr, Mb, Pb, Cr, Ni, Zn, and Br <p>CMB fitting parameters: 34 elements (Na to U), ions (NO₃⁻, SO₄²⁻, NH₄⁺, Na⁺, K⁺), carbon (OC, EC) were used to attribute PM₁₀ to 9 source types by CMB.</p> <p>Sensitivity tests: Conducted on 18 WTE profiles and 15 industrial profiles to assess the validity of RRRF source contributions.</p> <p>Goodness of fit: For all CMB runs, R² = 0.97±0.03, $\chi^2 = 0.79 \pm 0.49$, and % mass = 86.7±17.0%.</p> <p>Measurement/modeling uncertainties: Reported.</p> <p>Validation: The WTE source profile was used for apportionment prior to and after RRRF began operation to evaluate RRRF's contribution to PM₁₀ and toxic species.</p>	<p>Average CMB-calculated source contributions to PM₁₀ (in % of average mass from all 4 sites):</p> <table> <tr> <th>Source Type</th><th>FY96 (before RRRF began operation)</th><th>FY97 (after RRRF began operation)</th></tr> <tr> <td>Industry</td><td></td><td></td></tr> <tr> <td>Coal-fired power plant</td><td>1%</td><td>1%</td></tr> <tr> <td>WTE incinerator</td><td>1%</td><td>0%-2%</td></tr> <tr> <td>Steel plant</td><td>3%-4%</td><td>2%-3%</td></tr> <tr> <td>Transportation (vehicle exhaust)</td><td>13%-15%</td><td>12%-15%</td></tr> <tr> <td>Vegetative burning/meat cooking</td><td>6%-7%</td><td>7%-8%</td></tr> <tr> <td>Geological</td><td>17%-22%</td><td>16%-21%</td></tr> <tr> <td>Road salt</td><td>2%-6%</td><td>2%</td></tr> <tr> <td>Secondary ammonium sulfate</td><td>24%</td><td>21%-24%</td></tr> <tr> <td>Secondary ammonium nitrate</td><td>16%-19%</td><td>11%-13%</td></tr> <tr> <td>Average measured mass (µg/m³)</td><td>29.9±1.8 to 33.1±2.1</td><td>24.6±1.5 to 28.0±1.8</td></tr> <tr> <td>Number in average</td><td>135</td><td>136</td></tr> </table> <p>Contributions to PM₁₀ from RRRF's incinerator were undetectable. Contributions from RRRF sources to toxic species with profiles similar to target species were low, in the range of 1%-7%. Contributions from fossil fuel industries were low, but site- or area-specific profiles for coal-fired power plants, steel mills, petroleum refineries, ferrous alloy industries, and metal plating are needed for the midwest region of the U.S.</p>	Source Type	FY96 (before RRRF began operation)	FY97 (after RRRF began operation)	Industry			Coal-fired power plant	1%	1%	WTE incinerator	1%	0%-2%	Steel plant	3%-4%	2%-3%	Transportation (vehicle exhaust)	13%-15%	12%-15%	Vegetative burning/meat cooking	6%-7%	7%-8%	Geological	17%-22%	16%-21%	Road salt	2%-6%	2%	Secondary ammonium sulfate	24%	21%-24%	Secondary ammonium nitrate	16%-19%	11%-13%	Average measured mass (µg/m ³)	29.9±1.8 to 33.1±2.1	24.6±1.5 to 28.0±1.8	Number in average	135	136
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{*,†,‡}	Source Apportionment Method [‡]	Findings																																																																																																		
<p>Reference: Part of Project MOHAVE - <u>Measurement of Haze and Visual Effects</u>.^{bb}</p> <p>When: Twice daily, 12-hr (0700-1900 PST and 1900 to next day 0700 PST) sampling during 1992 with summer (July 12-Aug. 30) and winter (Jan. 15-Feb. 12) intensive studies.</p> <p>Where: At 40+ monitoring sites in and around Grand Canyon National Park. Only data from Marble Canyon and Indian Gardens are reported here.</p> <p>Ambient: Teflon, nylon, and quartz filter samples were acquired with IMPROVE samplers and analyzed for PM_{2.5} mass, elements (Na to Pb by PIXE), b_{abs} (by integrating sphere method), ions (NO₃, SO₄, and NH₄⁺ by IC), and carbon (OC and EC by TOR). Only elements and b_{abs} were used in this analysis.</p> <p>Source: No area-specific source profile measurements were taken. A new technique is being developed for verifying and extracting source profiles from ambient data sets. Selected trace element profiles for 14 metals are reported without associated uncertainties.</p>	<p>Solution: First, factor analysis was used to select trace elements to serve as independent variables for ordinary least squares regression modeling. Effective variance weighted least squares CMB7^b was used to apportion trace elements in the regression models to source types.</p> <p>Source profiles (from Pacific Northwest source profile library^{bc}):</p> <p>Industry (coal-fired power plant [NGS and MPP], copper smelter, paper mill, cement kiln, sulfite recovery boiler, aluminum processing, residual oil combustion, municipal incinerator, wood-fired boiler)</p> <p>Transportation (motor vehicles)</p> <p>Vegetative burning (agricultural burning, coniferous tree limb burns)</p> <p>Geological (soil dust)</p> <p>CMB fitting parameters: 14 elements and 8 source profiles were used in CMB attributions to 6 trace elements.</p> <p>Sensitivity tests: Not reported.</p> <p>Goodness of fit: Not reported.</p> <p>Measurement/modeling uncertainties: Only measurement uncertainties were reported.</p> <p>Validation: Singular value decomposition (SVD) of trace elements and sum equals one projection (SEOP) geometrical techniques were used to examine the validity of source profiles.</p>	<p>Average CMB-calculated source contributions to trace elements during the winter intensive study period:</p> <table><thead><tr><th>Source Type</th><th>Fe</th><th>Pb</th><th>Se</th><th>Br</th><th>As</th><th>K</th></tr></thead><tbody><tr><td>Industry</td><td></td><td></td><td></td><td></td><td></td><td></td></tr><tr><td>Coal-fired power plants</td><td>73%</td><td>0%</td><td>100%</td><td>0%</td><td>0%</td><td>15%</td></tr><tr><td>Copper smelter</td><td>0%</td><td>1%</td><td>0%</td><td>0%</td><td>100%</td><td>0%</td></tr><tr><td>Transportation</td><td>2%</td><td>92%</td><td>0%</td><td>18%</td><td>0%</td><td>0%</td></tr><tr><td>Vegetative burning</td><td>2%</td><td>6%</td><td>0%</td><td>82%</td><td>0%</td><td>79%</td></tr><tr><td>Geological</td><td>22%</td><td>0%</td><td>0%</td><td>0%</td><td>0%</td><td>5%</td></tr></tbody></table> <p>Indian Gardens</p> <table><thead><tr><th>Source Type</th><th>Fe</th><th>Pb</th><th>Se</th><th>Br</th><th>As</th><th>K</th></tr></thead><tbody><tr><td>Industry</td><td></td><td></td><td></td><td></td><td></td><td></td></tr><tr><td>Coal-fired power plants</td><td>32%</td><td>0%</td><td>98%</td><td>0%</td><td>0%</td><td>6%</td></tr><tr><td>Copper smelter</td><td>1%</td><td>36%</td><td>2%</td><td>0%</td><td>100%</td><td>0%</td></tr><tr><td>Transportation</td><td></td><td></td><td></td><td></td><td></td><td></td></tr><tr><td>Vegetative burning (coniferous tree limb burns and agricultural burning)</td><td>3%</td><td>8%</td><td>0%</td><td>91%</td><td>0%</td><td>81%</td></tr><tr><td>Geological</td><td>64%</td><td>0%</td><td>0%</td><td>0%</td><td>0%</td><td>13%</td></tr></tbody></table> <p>CMB analyses with adjusted profiles attributed ~90% of Se to coal-fired power plants, ~80% of Br to vegetative burning, and most of the As to copper smelters. Sulfur apportionment attributed ~50% of S to Se, linked to coal-fired power plants during summer and winter. During winter, ~50% of S and 3% of light extinction (b_{ext}) is related to emissions from vegetative burning. The combination of different receptor modeling techniques allowed source attribution to individual species with higher confidence.</p>	Source Type	Fe	Pb	Se	Br	As	K	Industry							Coal-fired power plants	73%	0%	100%	0%	0%	15%	Copper smelter	0%	1%	0%	0%	100%	0%	Transportation	2%	92%	0%	18%	0%	0%	Vegetative burning	2%	6%	0%	82%	0%	79%	Geological	22%	0%	0%	0%	0%	5%	Source Type	Fe	Pb	Se	Br	As	K	Industry							Coal-fired power plants	32%	0%	98%	0%	0%	6%	Copper smelter	1%	36%	2%	0%	100%	0%	Transportation							Vegetative burning (coniferous tree limb burns and agricultural burning)	3%	8%	0%	91%	0%	81%	Geological	64%	0%	0%	0%	0%	13%
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Study, Location, Period, and Measurements*†‡	Source Apportionment Method‡	Findings																																																																								
<p>Reference: Summer intensive study as part of Project MOHAVE - Measurement of <u>Haze</u> and <u>Visual Effects</u>.^{bd}</p> <p>When: Daily, 12-hr daytime (0700-1900 PST) and nighttime (1900 to next day 0700 PST) sampling during 1992 with summer (July 12-Aug. 30) and winter (Jan. 15-Feb. 12) intensive studies.</p> <p>Where: At 10 monitoring sites in and around Grand Canyon National Park. Summer data from the Meadview and Grand Canyon sites are reported here.</p> <p>Ambient: Teflon, quartz, and nylon filter samples were acquired with IMPROVE samplers. Samples were analyzed for PM_{2.5} mass, elements (Na to Pb by PIXE), b_{abs} (by integrating sphere method), ions (NO₃⁻, SO₄²⁻, and NH₄⁺ by IC), and carbon (OC and EC by TOR). HF and F samples were collected with hivol and denuder sampling systems and analyzed by fluoride-specific electrode. SAS particles were collected with slotted polyethylene impaction stages in hivol samplers and analyzed by SEM.</p> <p>Source: No area-specific source profile measurements were taken. Regional source profiles were developed using samples corresponding to air mass transport from a specific source region.^{be}</p>	<p>Solution: Second-generation CMB. This method assumes that separation of particles and SO₂ can occur in the MPP (Mojave power plant) plume during nighttime stable plume conditions.</p> <p>Source profiles:</p> <p>Industrial</p> <p>Mojave Power Project coal-fired power plant (MPP)^{be}</p> <p>Navajo Generating Station coal-fired power plant (NGS)^{be}</p> <p>Reid Gardner Generating Station coal-fired power plant (RGS)^{be}</p> <p>Central Utah coal-fired power plant (NH)^{be}</p> <p>Las Vegas urban area (LV)[†]</p> <p>Baja California, including Imperial Valley/northwest Mexico region (BC)[†]</p> <p>Northwest Nevada sources (NW1)[†]</p> <p>Local emissions near Grand Canyon (NW2)</p> <p>Arizona sources (AZ)[†]</p> <p>San Joaquin Valley sources (SJ)[†]</p> <p>South Coast air basin sources (LA)[†]</p> <p>southeast source region (SE)[†]</p> <p>Coal-fired power plant SO_x sources close to Petrified Forest National Park (AGS)[†]</p> <p>Colorado Plateau (CP)[†]</p> <p>Wasatch Front (WF)[†]</p> <p>† Regional profiles</p> <p>CMB fitting parameters: Not reported.</p> <p>Sensitivity tests: Not reported.</p> <p>Goodness of fit: Not reported.</p> <p>Measurement/modeling uncertainties: Not reported.</p> <p>Validation: None</p>	<p>Average CMB-calculated source contributions to average summertime SO₄²⁻ and SO_x:</p> <table><thead><tr><th colspan="2">SO₄²⁻</th><th colspan="2">SO_x (SO₂ plus particulate SO₄²⁻)</th></tr><tr><th>Regional Sources</th><th>Meadview</th><th>Hopi Point</th><th>Meadview</th></tr></thead><tbody><tr><td>MPP</td><td>5.5%</td><td>4.3%</td><td>39.3%</td></tr><tr><td>NGS</td><td>0%</td><td>0.8%</td><td>0%</td></tr><tr><td>RGS</td><td>0%</td><td>0%</td><td>0%</td></tr><tr><td>LV</td><td>38.6%</td><td>18.6%</td><td>26.9%</td></tr><tr><td>BC</td><td>11.6%</td><td>42.1%</td><td>5.9%</td></tr><tr><td>NW1</td><td>0%</td><td>0.6%</td><td>0%</td></tr><tr><td>NW2</td><td>2.4%</td><td>0.1%</td><td>1.8%</td></tr><tr><td>AZ</td><td>3.3%</td><td>0.1%</td><td>1.6%</td></tr><tr><td>SJ</td><td>13.5%</td><td>17.5%</td><td>7.6%</td></tr><tr><td>LA</td><td>17.3%</td><td>14.4%</td><td>11.9%</td></tr><tr><td>SE</td><td></td><td>4.3%</td><td></td></tr><tr><td>AGS</td><td></td><td>0.1%</td><td></td></tr><tr><td>SE and AGS</td><td></td><td></td><td>5.0%</td></tr><tr><td>Excess</td><td>7.7%</td><td>1.6%</td><td></td></tr><tr><td>Not explained</td><td></td><td></td><td>4.6%</td></tr><tr><td></td><td></td><td></td><td>11.7%</td></tr></tbody></table> <p>Straightforward CMB application on the Project MOHAVE data set using regional profiles resulted in significant underestimation of total sulfate oxides (SO_x, sum of SO₂ and particulate SO₄²⁻). Second-generation CMB reported that MPP was the major source of SO_x and the Las Vegas urban area was the major source of SO₄²⁻ at the Meadview site. Baja California contributed a majority of SO_x and SO₄²⁻ at the Grand Canyon site.</p>	SO ₄ ²⁻		SO _x (SO ₂ plus particulate SO ₄ ²⁻)		Regional Sources	Meadview	Hopi Point	Meadview	MPP	5.5%	4.3%	39.3%	NGS	0%	0.8%	0%	RGS	0%	0%	0%	LV	38.6%	18.6%	26.9%	BC	11.6%	42.1%	5.9%	NW1	0%	0.6%	0%	NW2	2.4%	0.1%	1.8%	AZ	3.3%	0.1%	1.6%	SJ	13.5%	17.5%	7.6%	LA	17.3%	14.4%	11.9%	SE		4.3%		AGS		0.1%		SE and AGS			5.0%	Excess	7.7%	1.6%		Not explained			4.6%				11.7%
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<p>Reference: Mt. Zirkel Visibility Study.^{bf}</p> <p>When: 12-hr sampling (0600-1800 MST) was conducted for a one-year period from Dec. 1, 1994, to Nov. 30, 1995. In addition, 6-hr, 2 times/day (0600-1200 and 1200-1800 MST) sampling was conducted during three seasonal intensive periods (winter [Feb. 6-Mar. 2], summer [Aug. 3-Sept. 3], and fall [Sept. 15-Oct. 15]) in 1995.</p> <p>Where: Mt. Zirkel Wilderness Area, Colorado, at 5 sites in Colorado (Buffalo Pass, Gilpin Creek, Juniper Mountain, Hayden VOR, and Hayden wastewater) and one site in Wyoming (Baggs).</p> <p>Ambient: PM_{2.5} samples were acquired with SFS for mass, <i>b</i>_{abs} (by densitometry), 40 elements (Na to U by XRF), ions (Cl⁻, NO₃⁻, and SO₄²⁻ by IC; NH₄⁺ by AC; and K⁺ by AAS), carbon (OC and EC by TOR), and sulfur isotopes (³²S and ³⁴S by accelerator mass spectrometry).</p> <p>Gaseous SO₂ samples were acquired with potassium carbonate impregnated cellulose filters.</p> <p>Gaseous NH₃ and HNO₃ were acquired by denuder difference with SGS during intensive periods.</p> <p>Source: Source profiles were derived from 10 coal-fired power plant, 2 residential coal combustion, 3 residential wood combustion, 4 wildfire, 5 geothermal hot spring, 9 roadside vehicle exhaust, and 12 geological samples. Source profiles applied in the study are reported with associated uncertainties in Watson et al.^{aw}</p>	<p>Solution: Effective variance weighted least squares CMB7.^b).</p> <p>Source profiles:</p> <p>Industry (Craig and Hayden coal-fired power plants burning bituminous coal)</p> <p>Transportation (roadside sampling near major highways, diesel bus terminal)</p> <p>Vegetative burning (residential wood combustion from fireplace and woodstove, wildfire of pinion and juniper)</p> <p>Geological (paved road dust, unpaved road dust, soil)</p> <p>Secondary aerosol (ammonium sulfate, ammonium nitrate)</p> <p>Others (residential coal combustion, geothermal hot springs, regional background sources)</p> <p>CMB fitting parameters: 21 elements (Na to U), ions (Cl⁻, NO₃⁻, SO₄²⁻, and K⁺), carbon (OC and EC), and gaseous SO₂ from 367 samples were used in CMB attributions to 6 source types.</p> <p>Sensitivity tests: Reported with 7 trial runs.</p> <p>Goodness of fit: Reported for individual CMB output in Appendix D of Watson et al.^{bf}</p> <p>Measurement/modeling uncertainties: Reported.</p> <p>Validation: Dispersion modeling was used to verify CMB-calculated source contributions to PM_{2.5} mass and visibility.</p>	<p>Average CMB-calculated source contributions to PM_{2.5} (in % mass):</p> <table><tr><th>Source Type</th><th>Buffalo Pass</th><th>Gilpin Creek</th><th>Juniper Mountain</th><th>Hayden VOR</th><th>Hayden Waste-water</th><th>Baggs</th></tr><tr><td>Industry</td><td>4.4%</td><td>2.0%</td><td>1.6%</td><td>15.0%</td><td>15.4%</td><td>4.8%</td></tr><tr><td>Transportation</td><td>19.3%</td><td>32.2%</td><td>31.1%</td><td>27.6%</td><td>30.2%</td><td>27.6%</td></tr><tr><td>Vegetative burning</td><td>18.5%</td><td>18.4%</td><td>17.9%</td><td>11.3%</td><td>10.5%</td><td>15.0%</td></tr><tr><td>Geological</td><td>29.1%</td><td>15.1%</td><td>19.3%</td><td>24.2%</td><td>16.1%</td><td>21.6%</td></tr><tr><td>Secondary ammonium sulfate</td><td>24.7%</td><td>26.3%</td><td>26.9%</td><td>17.2%</td><td>19.9%</td><td>27.6%</td></tr><tr><td>Secondary ammonium nitrate</td><td>4.0%</td><td>5.9%</td><td>3.2%</td><td>4.7%</td><td>7.9%</td><td>3.5%</td></tr><tr><td>Average calculated PM_{2.5} mass (µg/m³)</td><td>4.7</td><td>4.3</td><td>4.5</td><td>5.6</td><td>5.4</td><td>3.8</td></tr><tr><td>Number in average</td><td>64</td><td>47</td><td>47</td><td>62</td><td>66</td><td>57</td></tr></table> <p>Substantial differences were found among CALMET/CALPUFF, CMB, and continuous measurements with the greatest disagreement when transformation in fogs and clouds occurred.</p> <p>CALMET/CALPUFF plume chemistry modeling often underestimated measured PM_{2.5} and light extinction. This discrepancy may be due to: 1) source contributions from outside of emission domain, 2) inaccurate emission estimates for intermittent sources during episodes, and 3) inadequate mechanism for determining aqueous-phase conversion of SO₂ to sulfate that are the cause of most event with perceptible visibility impairment.</p>	Source Type	Buffalo Pass	Gilpin Creek	Juniper Mountain	Hayden VOR	Hayden Waste-water	Baggs	Industry	4.4%	2.0%	1.6%	15.0%	15.4%	4.8%	Transportation	19.3%	32.2%	31.1%	27.6%	30.2%	27.6%	Vegetative burning	18.5%	18.4%	17.9%	11.3%	10.5%	15.0%	Geological	29.1%	15.1%	19.3%	24.2%	16.1%	21.6%	Secondary ammonium sulfate	24.7%	26.3%	26.9%	17.2%	19.9%	27.6%	Secondary ammonium nitrate	4.0%	5.9%	3.2%	4.7%	7.9%	3.5%	Average calculated PM _{2.5} mass (µg/m ³)	4.7	4.3	4.5	5.6	5.4	3.8	Number in average	64	47	47	62	66	57
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{*†‡}	Source Apportionment Method [‡]	Findings
Reference: REVEAL regional visibility experiment assessment. ^{bg} When: Daily 24-hr sampling during July and August 1993. Where: Two sites (Chilliwack [in the eastern valley] and Pitt Meadows) in the Lower Fraser Valley, BC, Canada. Ambient: PM _{2.5} samples on Teflon, quartz, and nylon filters were acquired with IMPROVE samplers and analyzed for mass, elements (Na to Pb by PIXE), ions (NO ₃ ⁻ , SO ₄ ²⁻ , and NH ₄ ⁺ by IC), and carbon (OC and EC by TOR).	Solution: Effective variance weighted least squares CMB7. ^b Source profiles: Industry (coal- and oil-fired power plants [^] , incinerator [^] , paper mill #23103) ^{ac} , Transportation (vehicle emissions from dynamometer tests) ^{c-e} Vegetative burning (residential wood combustion) ^{ea} Geological (paved road dust, soil) ^{bh} Marine aerosol ^f Secondary aerosol (ammonium sulfate, ammonium nitrate) ^g [^] Only used for CMB tests CMB fitting parameters: 25 elements (Na to Pb), ions (NO ₃ ⁻ , SO ₄ ²⁻ , NH ₄ ⁺), and carbon (OC, EC) from 54 samples were used in CMB attributions to 6 source types. Sensitivity tests: Reported with 7 trial runs. Goodness of fits: Not reported. Measurement/modeling uncertainties: Reported. Validation: CMB calculations were compared with emissions inventory. ^{bi}	Average CMB-calculated source contributions to PM _{2.5} (in % mass): Source Type Transportation Vegetative burning Geological Marine aerosol Secondary ammonium sulfate Secondary ammonium nitrate Average Measured PM _{2.5} Mass (μg/m ³) Number in Average Chilliwack 34% 8% 3% 3% 25% 27% 9.8 ± 0.4 28 Pitt Meadows 43% 9% 5% 5% 27% 12% 8.1 ± 0.3 26 Emissions Inventory 20% — 58% — — — — —

While paper mill contributions to PM_{2.5} were insignificant, its contributions to secondary ammonium sulfate may be significant and need to be evaluated.

Secondary ammonium nitrate levels in the eastern valley of the Chilliwack site were 2 to 3 times those at the Pitt Meadows site.

Large discrepancies were found between CMB-calculated source contributions and those estimated in the emissions inventory for transportation and geological.

Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																																																																																					
<p>Reference:</p> <p>Panel on Energy Research and Development (PERD) project.^{bj}</p> <p>When: Daily, 24-hr (starting at 1000 EST) sampling for 10-day periods during spring (March/April) and summer (July) 1998.</p> <p>Where: Ontario, Canada, at the urban Evans site (a very high traffic area) in Toronto and at the rural Egbert (CARE) site 150 km north of urban Toronto.</p> <p>Ambient: Teflon and quartz filter samples were acquired with Partisol, VAPS, dichotomous, and IMPROVE samplers. Samples were analyzed for PM_{2.5} mass, 23 elements (Na to U by XRF and PIXE), 5 ions (Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, and NH₄⁺ by IC/CE), carbon (OC and EC by TOR, PESA, and LIPM), organics (12 organic acids, n-alkanes, and PAHs by GC/MS).</p> <p>Source: No area-specific source profile measurements were taken. Source profiles applied are reported (without associated uncertainties) in Appendix B of Brook et al.^{bj}</p>	<p>Solution: Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <p>Industry (coal-fired power plants^{aw}, petroleum^{ae}, metal processing/steel plant^{ae}, smelter^{ae}, tire incineration^{ae})</p> <p>Transportation (gas and diesel, motor vehicle dynamometer)^{c,d}</p> <p>Vegetative burning (residential wood combustion from fireplaces and woodstoves)^{ba}</p> <p>Geological (road dust, agricultural soil, limestone)^{az}</p> <p>Secondary aerosol (ammonium sulfate, ammonium nitrate, organics)^e</p> <p>CMB fitting parameters: 22 elements (Na to U), ions (Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺), and carbon (OC, EC) from 42 samples were used in CMB attributions to 11 source types. 22 out of 58 tested profiles were selected.</p> <p>Sensitivity tests: Reported in Appendix A of Brook et al.^{bk}</p> <p>Goodness of fits: Seasonal and site average for all CMB runs, R² = 0.68 to 0.85, χ^2 = 1.9 to 3.7, and % mass = 93.6±28.2 to 128.4±28.5.</p> <p>Measurement/model uncertainties: Not reported.</p> <p>Validation: Backward trajectory was used to determine air mass transport.</p>	<p>Average CMB-calculated source contributions to PM_{2.5} (in % mass):</p> <table><tr><th>Source Type</th><th>Evans (Toronto) urban site Mar/Apr '98</th><th>July '98</th><th>Egbert (CARE) semi-rural site Mar/Apr '98</th><th>July '98</th></tr><tr><td>Industry</td><td></td><td></td><td></td><td></td></tr><tr><td>Coal-fired power plant</td><td><1%</td><td>3%</td><td>2%</td><td>7%</td></tr><tr><td>Metal processing/steel plant</td><td>1%</td><td>7%</td><td>3%</td><td><1%</td></tr><tr><td>Smelter</td><td><1%</td><td>1%</td><td><1%</td><td>3%</td></tr><tr><td>Tire incinerator</td><td><1%</td><td><1%</td><td>4%</td><td><1%</td></tr><tr><td>Transportation</td><td>37%</td><td>63%</td><td>26%</td><td>14%</td></tr><tr><td>Vegetative Burning (RWC)</td><td>16%</td><td>4%</td><td>14%</td><td>15%</td></tr><tr><td>Geological</td><td></td><td></td><td></td><td></td></tr><tr><td>Paved road dust</td><td>1%</td><td>1%</td><td><1%</td><td><1%</td></tr><tr><td>Agricultural soil</td><td>11%</td><td>2%</td><td>6%</td><td>1%</td></tr><tr><td>Limestone</td><td><1%</td><td><1%</td><td>1%</td><td><1%</td></tr><tr><td>Secondary ammonium sulfate</td><td>26%</td><td>19%</td><td>33%</td><td>32%</td></tr><tr><td>Secondary ammonium nitrate</td><td>9%</td><td><1%</td><td>10%</td><td>1%</td></tr><tr><td>Secondary organics</td><td><1%</td><td><1%</td><td>5%</td><td>27%</td></tr><tr><td>Average Measured PM_{2.5} Mass (µg/m³)</td><td>13.9</td><td>12.4</td><td>9.3</td><td>6.6</td></tr><tr><td>Number in Average</td><td>8</td><td>11</td><td>13</td><td>10</td></tr></table> <p>Vehicle exhaust was the largest contributor (37%-63%) to PM_{2.5} at the urban site. Secondary ammonium sulfate was the largest contributor (32%-33%) at the semi-rural Egbert site.</p> <p>Source contributions from industry and fossil fuel use from Ontario and upwind areas may contribute to elevated sulfate concentrations which varied from 1% to 11%, with 2%-7% derived from coal-fired power plants.</p> <p>Organic speciation (acids and alkanes) explains only 1%-10% of total organics. Biogenic sources (odd-number alkanes >> even-number alkanes) may have contributed to primary and secondary organic aerosol at the semi-rural site.</p>	Source Type	Evans (Toronto) urban site Mar/Apr '98	July '98	Egbert (CARE) semi-rural site Mar/Apr '98	July '98	Industry					Coal-fired power plant	<1%	3%	2%	7%	Metal processing/steel plant	1%	7%	3%	<1%	Smelter	<1%	1%	<1%	3%	Tire incinerator	<1%	<1%	4%	<1%	Transportation	37%	63%	26%	14%	Vegetative Burning (RWC)	16%	4%	14%	15%	Geological					Paved road dust	1%	1%	<1%	<1%	Agricultural soil	11%	2%	6%	1%	Limestone	<1%	<1%	1%	<1%	Secondary ammonium sulfate	26%	19%	33%	32%	Secondary ammonium nitrate	9%	<1%	10%	1%	Secondary organics	<1%	<1%	5%	27%	Average Measured PM _{2.5} Mass (µg/m ³)	13.9	12.4	9.3	6.6	Number in Average	8	11	13	10
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																																										
<p>Reference: Mexico City PM_{2.5} study.^{bl}</p> <p>When: Two 12-hr (8am to 8pm) samples every third day from December 1989 to February 1990.</p> <p>Where: Mexico City, Mexico, in two medium-income neighborhoods 5 km away from downtown Mexico City and ~0.5 km away from major traffic roads.</p> <p>Ambient: Teflon, quartz, and nylon filters samples were acquired with low-volume (12 L/min) single- and dual-filter-pack systems and preceded with Teflon-coated glass PM_{2.5} cyclone and two annular denuders. Samples were analyzed for mass, elements (Na to Pb by PIXE), ions (F⁻, Cl⁻, NO₃⁻, and SO₄²⁻ by IC), and carbon (OC and EC by TOT).</p> <p>Source: Developed an area-specific source profile for Mexico City soil by grab sampling followed by chemical analysis. This source profile was not reported.</p>	<p>Solution: Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:^{ac}</p> <p>Industry (oil refinery^{**}, cement plant, smelter)</p> <p>Transportation (non-catalyst gasoline vehicles and heavy-duty diesel vehicles)^{††}</p> <p>Geological (local soil and California soils from Los Angeles and Riverside)</p> <p>Secondary aerosol (sulfate and nitrate).</p> <p>^{**} Oil refinery profile was adjusted to reflect "18 de Marzo" refinery process.</p> <p>^{††} Gasoline profile was corrected to reflect 1989 Mexican gasoline lead content.</p> <p>CMB fitting parameters: 18 elements (Na to Pb), 3 ions (F⁻, NO₃⁻, SO₄²⁻), and carbon (OC, EC) from 33 samples were used in CMB attributions to 9 source types.</p> <p>Sensitivity tests: None.</p> <p>Goodness of fits: Comparison of model calculations to measured mass are reported.</p> <p>Measurement/modeling uncertainties: Not reported.</p> <p>Validation: None.</p>	<p>Average source contributions to PM_{2.5} (in % mass, estimated from the figures):</p> <table> <tr> <th>Source Type</th><th>Daytime (8am to 8pm)</th><th>Nighttime (8pm to next day 8am)</th></tr> <tr> <td>Industry</td><td></td><td></td></tr> <tr> <td>Oil refinery</td><td>12%</td><td>17%</td></tr> <tr> <td>Cement plant</td><td>0.5%</td><td>1%</td></tr> <tr> <td>Smelter</td><td>1.5%</td><td>2%</td></tr> <tr> <td>Transportation</td><td></td><td></td></tr> <tr> <td>Non-catalyst gasoline vehicles</td><td>41%</td><td>36%</td></tr> <tr> <td>Heavy-duty diesel vehicles</td><td>6%</td><td>4%</td></tr> <tr> <td>Geological (combination of Mexico and California soil)</td><td>19%</td><td>20%</td></tr> <tr> <td>Secondary sulfate</td><td>9%</td><td>12%</td></tr> <tr> <td>Secondary nitrate</td><td>5%</td><td>3%</td></tr> <tr> <td>Secondary organics</td><td>4%</td><td>2.5%</td></tr> <tr> <td>Average Measured PM_{2.5} Mass (µg/m³)</td><td>118.9</td><td>65.8</td></tr> <tr> <td>Number in Average</td><td>19</td><td>14</td></tr> </table> <p>Daytime and nighttime concentrations varied by twofold with similar relative source contributions. The largest contributors are non-catalyst gasoline vehicles (36%-41%), geological dust (~20%), and secondary aerosol (~20%). Fossil fuel industry (mainly oil refinery) contributed 14%-20% of PM_{2.5}.</p> <p>CMB results will be verified as control actions are implemented in the Mexico City metropolitan area:</p> <ul style="list-style-type: none"> – Shutdown of the 18 de Marzo refinery, – Use of unleaded gasoline (magna sin) with catalytic converters began in 1991, – Lead content reduced in leaded gasoline, and – Reactive HC content reduced in leaded gasoline. 	Source Type	Daytime (8am to 8pm)	Nighttime (8pm to next day 8am)	Industry			Oil refinery	12%	17%	Cement plant	0.5%	1%	Smelter	1.5%	2%	Transportation			Non-catalyst gasoline vehicles	41%	36%	Heavy-duty diesel vehicles	6%	4%	Geological (combination of Mexico and California soil)	19%	20%	Secondary sulfate	9%	12%	Secondary nitrate	5%	3%	Secondary organics	4%	2.5%	Average Measured PM _{2.5} Mass (µg/m ³)	118.9	65.8	Number in Average	19	14
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Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,t,t}	Source Apportionment Method [†]	Findings																																																								
<p>Reference: Czech air quality study.^{bm}</p> <p>When: 24-hr sampling during winter and summer 1993 and winter 1994.</p> <p>Where: Teplice (an industrial city 90 km northwest of Prague) and Prachatice (a residential town 130 km south of Prague), Czech Republic.</p> <p>Ambient: Teflon and quartz filter samples were acquired with VAPS samplers. Samples were analyzed for PM_{2.5} mass, elements (by XRF), carbon (OC and EC by TOA), and 12 PAHs (by GC/MS with Method TO-13^{bm}). PM_{coarse} samples underwent single particle analysis by SEM/EDX. Gaseous SO₂, HCl, HNO₃, and HF were collected with annular denuders and analyzed by IC.</p> <p>Source: Source samples were acquired by DSP from 4 lignite-fueled coal-fired power plants, glass factory, hospital incinerator, hand-fired coal stove, lignite-fueled residential space heating furnace, and roadway tunnel (90% gasoline/10% diesel-fueled vehicles). Source profiles used in CMB calculations are reported without associated uncertainties.</p>	<p>Solution: Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <p>Industry (Ledvice power plant, hospital incinerator)</p> <p>Transportation (motor vehicle exhaust)</p> <p>Home heating (smoldering and high-temperature phases)</p> <p>Secondary sulfate</p> <p>CMB fitting parameters: 18 elements, ion (SO₄), and carbon (OC, EC) were used in CMB attributions to as many as 6 source types.</p> <p>Sensitivity tests: Not reported.</p> <p>Goodness of fit: Reported χ^2 of 0.5 to 1.8.</p> <p>Measurement/modeling uncertainties: Measurement uncertainties were reported.</p> <p>Validation: CMB-calculated source contributions to PM_{2.5} and to total sulfur (SO₂ plus SO₄) were compared with emissions inventory estimates. Annual emission rates for SO₂ from power plants were scaled down by a factor of 4 to compare with winter period SO₂ emissions from home heating. Summer and winter ratios of B(a)P to Pb, As to PM_{2.5} mass, and PAH to PM_{2.5} mass were examined to identify residential space heating emissions during winter.</p>	<p>Average CMB-calculated source contributions to PM_{2.5} at the Teplice site (in % mass):</p> <table><tr><th>Source Type</th><th>1st episode</th><th>2nd episode</th><th>Average winter 1993</th></tr><tr><td>Industry</td><td></td><td></td><td></td></tr><tr><td>Power plants</td><td>1.8%</td><td>11.4%</td><td>3.3%</td></tr><tr><td>Incinerator</td><td>3.4%</td><td>4.5%</td><td>6.9%</td></tr><tr><td>Transportation (vehicle exhaust)</td><td>1.6%</td><td>2.5%</td><td>2.8%</td></tr><tr><td>Residential heating (smoldering coal)</td><td>17.5%</td><td>18.8%</td><td>24.0%</td></tr><tr><td>Residential heating (high-temperature coal)</td><td>1.6%</td><td>4.3%</td><td><1.0%</td></tr><tr><td>Secondary sulfate</td><td>48%</td><td>50%</td><td>40%</td></tr><tr><td>Unexplained</td><td>25%</td><td>8.5%</td><td>23%</td></tr><tr><td>Average measured PM_{2.5} mass (µg/m³)</td><td></td><td></td><td>51.1±2.8</td></tr><tr><td>Number in average</td><td></td><td></td><td>117</td></tr></table> <p>Average CMB-calculated source contributions to total sulfur (SO₂ plus SO₄) (in µg/m³):</p> <table><tr><th>Source Type</th><th>1st episode</th><th>2nd episode</th><th>Average winter 1993</th></tr><tr><td>Total sulfur (home heating)</td><td>50</td><td>24</td><td>40</td></tr><tr><td>Total sulfur (power plants)</td><td>50</td><td>76</td><td>60</td></tr></table> <p>Power plants and residential heating contributions to total sulfur were scaled to receptor modeling results using ratios of SO₂ to PM_{2.5} emissions. SEM results and past studies showed that SO₂ converts to SO₄ by a heterogeneous mechanism. Coal-fired power plants were found to be a significant source of SO₂ and SO₄.</p> <p>Approximately 85% of PM_{2.5} analyzed by SEM was sulfate or carbonaceous particles. The remaining PM_{2.5} consisted of fly ash from power plants or home heating. PM_{coarse} levels were 10%-30% of PM_{2.5} levels. PM_{coarse} was dominated by fly ash and soil.</p>	Source Type	1st episode	2nd episode	Average winter 1993	Industry				Power plants	1.8%	11.4%	3.3%	Incinerator	3.4%	4.5%	6.9%	Transportation (vehicle exhaust)	1.6%	2.5%	2.8%	Residential heating (smoldering coal)	17.5%	18.8%	24.0%	Residential heating (high-temperature coal)	1.6%	4.3%	<1.0%	Secondary sulfate	48%	50%	40%	Unexplained	25%	8.5%	23%	Average measured PM _{2.5} mass (µg/m ³)			51.1±2.8	Number in average			117	Source Type	1st episode	2nd episode	Average winter 1993	Total sulfur (home heating)	50	24	40	Total sulfur (power plants)	50	76	60
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Table 1. (Continued)

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Reference: Source apportionment study in Qalabotjha, South Africa. ^{bo} When: Daily, 24-hr sampling (noon to noon) was conducted during winter (June 21-July 30) 1997. Where: Qalabotjha (150 km southeast of Johannesburg), South Africa, at 4 sites. Sampling was done at 3 urban sites (Police Station, Crèche, and Clinic) in a black township of Qalabotjha and at 1 transport site (Bowling Club) in Villiers, a white community southwest of Qalabotjha. Ambient: Teflon and quartz filter samples were acquired with PM ₁₀ SA-246 size-selective inlet and PM _{2.5} URG cyclone sampling systems. Samples were analyzed for 40 elements (Na to U by XRF), ions (Cl ⁻ , NO ₃ ⁻ , and SO ₄ ²⁻ by IC; NH ₄ ⁺ by AC; and Na ⁺ and K ⁺ by AAS), and carbon (4 OC fractions [OC1, OC2, OC3, OC4], 3 EC fractions [EC1, EC2, EC3], and pyrolyzed carbon [OPTC]). ^{ch} Source: A total of 28 source profiles for residential coal and low-smoke fuels combustion, coal-fired power plant, sinter plant, lime kiln, gasoline vehicle exhaust, biomass burning, and geological material were acquired. Source profiles with associated uncertainties are reported.	Solution: Effective variance weighted least squares CMB7. ^b Source profiles: Industry (coal-fired power plant fly ash, metallurgical sinter plant, lime kiln) Transportation (leaded gasoline emissions from pickup truck) Biomass burning (kindled grass fire, domestic coal, low-smoke fuels, cooking) Geological (paved road dust, soil) Secondary aerosol (ammonium sulfate, ammonium nitrate) ^e CMB fitting parameters: 13 elements (Na to U), ions (Cl ⁻ , NO ₃ ⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , Na ⁺ , K ⁺), and carbon (OC1, OC2, OC3, OC4, OPTC, EC1, EC2, and EC3) from 120 samples were used in CMB attributions to 11 source types. Sensitivity tests: Reported in Engelbrecht and Swanepoel. ^{bp} Goodness of fit: Reported in Appendix Q of Engelbrecht and Swanepoel. ^{bp} Measurement/modeling uncertainties: Reported. Validation: CMB results from period 1 (days 1-10, burning regular D-grade domestic coal), period 2 (days 11-20, burning almost only low-smoke fuels with some D-grade coal), and period 3 (days 21-30 burning only low-smoke fuels) were compared to evaluate the effectiveness of low-smoke fuels on ambient air quality. Analysis of variance and dispersion modeling were used to verify pollution transport. ^{bq}	Source contributions to average PM _{2.5} at the 3 urban Qalabotjha sites (in % mass): <table><tr><th>Source Type</th><th>Period 1 (burning only D-grade coal)</th><th>Period 2 (burning mix of D-grade coal and low-smoke fuels)</th><th>Period 3 (burning only low-smoke fuels)</th></tr><tr><td>Industry</td><td></td><td></td><td></td></tr><tr><td>Coal-fired power plant</td><td>0.1%</td><td>0.2%</td><td>0.2%</td></tr><tr><td>Arc furnace</td><td>0.5%</td><td>0.3%</td><td>0.4%</td></tr><tr><td>Transportation</td><td></td><td></td><td></td></tr><tr><td>Leaded vehicle exhaust</td><td>0.1%</td><td>0.1%</td><td>0.1%</td></tr><tr><td>Vegetative burning/heating and cooking</td><td>12.8%</td><td>16.0%</td><td>12.5%</td></tr><tr><td>Biomass</td><td>60.1%</td><td>62.7%</td><td>63.7%</td></tr><tr><td>Residential coal combustion</td><td></td><td></td><td></td></tr><tr><td>Geological</td><td></td><td></td><td></td></tr><tr><td>Soil</td><td>1.2%</td><td>0.6%</td><td>1.2%</td></tr><tr><td>Lime</td><td>0.2%</td><td>0.1%</td><td>0.1%</td></tr><tr><td>Potassium nitrate</td><td>1.2%</td><td>0.5%</td><td>0.6%</td></tr><tr><td>Ammonium chloride</td><td>0.1%</td><td>0.3%</td><td>0.3%</td></tr><tr><td>Secondary ammonium sulfate</td><td>6.0%</td><td>6.5%</td><td>5.8%</td></tr><tr><td>Secondary ammonium nitrate</td><td>1.1%</td><td>1.3%</td><td>1.8%</td></tr><tr><td>Unexplained</td><td>18%</td><td>11.3%</td><td>13.2%</td></tr><tr><td>Average Measured PM_{2.5} Mass (µg/m³)</td><td>126</td><td>101</td><td>112</td></tr><tr><td>Number in average</td><td>14</td><td>8</td><td>8</td></tr></table> Average PM _{2.5} and PM ₁₀ mass decreased by 20% and 25%, respectively, from the D-grade coal combustion period (days 1 to 10) to the mostly low-smoke fuels combustion period (days 11-20) with similar source contributions. Modeling of the continuous mass data showed that air quality improved by more than 60% when low-smoke fuels were used. This experience demonstrates the potential utility of using alternative domestic cooking and heating fuels as a means of pollution control.	Source Type	Period 1 (burning only D-grade coal)	Period 2 (burning mix of D-grade coal and low-smoke fuels)	Period 3 (burning only low-smoke fuels)	Industry				Coal-fired power plant	0.1%	0.2%	0.2%	Arc furnace	0.5%	0.3%	0.4%	Transportation				Leaded vehicle exhaust	0.1%	0.1%	0.1%	Vegetative burning/heating and cooking	12.8%	16.0%	12.5%	Biomass	60.1%	62.7%	63.7%	Residential coal combustion				Geological				Soil	1.2%	0.6%	1.2%	Lime	0.2%	0.1%	0.1%	Potassium nitrate	1.2%	0.5%	0.6%	Ammonium chloride	0.1%	0.3%	0.3%	Secondary ammonium sulfate	6.0%	6.5%	5.8%	Secondary ammonium nitrate	1.1%	1.3%	1.8%	Unexplained	18%	11.3%	13.2%	Average Measured PM _{2.5} Mass (µg/m ³)	126	101	112	Number in average	14	8	8
Source Type	Period 1 (burning only D-grade coal)	Period 2 (burning mix of D-grade coal and low-smoke fuels)	Period 3 (burning only low-smoke fuels)																																																																											
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Soil	1.2%	0.6%	1.2%																																																																											
Lime	0.2%	0.1%	0.1%																																																																											
Potassium nitrate	1.2%	0.5%	0.6%																																																																											
Ammonium chloride	0.1%	0.3%	0.3%																																																																											
Secondary ammonium sulfate	6.0%	6.5%	5.8%																																																																											
Secondary ammonium nitrate	1.1%	1.3%	1.8%																																																																											
Unexplained	18%	11.3%	13.2%																																																																											
Average Measured PM _{2.5} Mass (µg/m ³)	126	101	112																																																																											
Number in average	14	8	8																																																																											

Table 1. (Continued)

Study, Location, Period, and Measurements*†‡	Source Apportionment Method†	Findings																																																									
<p>Reference: PM_{2.5} study in the Sihwa area, Korea.</p> <p>When: 24-hr sampling (starting at 0900) during 4 intensive periods in 1998 (Feb. 16-25, June 8-18, Aug. 24-Sept. 2, Nov. 30-Dec. 9) and 1 intensive period in 1999 (Jan. 28-Feb. 11).</p> <p>Where: Sihwa, Korea (~30 km south of Seoul, in a residential area near a heavy industry complex).</p> <p>Ambient: 24-hr Teflon, quartz, and nylon filter samples were acquired with dichotomous and URG samplers. Samples were analyzed for mass, 22 elements (Na to Pb by ICP-MS), ions (Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺, Na⁺, K⁺, Mg⁺⁺, and Ca⁺⁺ by IC), and carbon (OC and EC by TMO). Gaseous precursors were acquired for HNO₃, HCl, and NH₃ (not used for CMB calculations).</p> <p>Source: No area-specific source profile measurements were taken.</p>	<p>Solution: Background trajectory analysis and factor analysis were used to identify source types for CMB analysis. Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <p>Industry (heavy oil combustion #13502, incinerator #17105, copper smelter #29209, metallurgy #28305)^{ae}</p> <p>Transportation (diesel exhaust only #32203)^{ae}</p> <p>Vegetative burning (agricultural burning #42304)^{ae}</p> <p>Geological material (soil and road dust #41120, construction #43304)</p> <p>Marine aerosol (#43101)^{ad}</p> <p>Secondary aerosol (ammonium sulfate, ammonium nitrate)^g</p> <p>Non-vehicle OC</p> <p>CMB fitting parameters: 18 elements (Na to Pb), ions (SO₄²⁻, NO₃⁻, NH₄⁺), and carbon (OC, EC) from 53 samples were used in CMB attributions to 12 source types.</p> <p>Sensitivity tests: Not reported.</p> <p>Goodness of fit: For all CMB runs, average R² = 0.98, $\chi^2 = 1.72$, and % mass = 92.6%.</p> <p>Measurement/modeling uncertainties: Reported.</p> <p>Validation: Backward trajectory was used to evaluate air mass transport.</p>	<p>Average CMB-calculated source contributions to PM_{2.5} (in % mass):</p> <table> <tr> <th>Source Type</th><th>Average SCE of 5 intensives</th><th>Average over SW and W wind</th></tr> <tr> <td>Industry</td><td></td><td></td></tr> <tr> <td>Heavy oil combustion</td><td>0.3%</td><td>0.7%</td></tr> <tr> <td>Incinerator</td><td>4.9%</td><td>6.1%</td></tr> <tr> <td>Metallurgy</td><td>0.8%</td><td>0.8%</td></tr> <tr> <td>Copper industry</td><td>0.5%</td><td>0.7%</td></tr> <tr> <td>Transportation (diesel exhaust)</td><td>8.6%</td><td>7.2%</td></tr> <tr> <td>Vegetative burning (agricultural burning)</td><td>11.3%</td><td>10.5%</td></tr> <tr> <td>Geological</td><td></td><td></td></tr> <tr> <td>Road dust</td><td>6.4%</td><td>5.0%</td></tr> <tr> <td>Construction</td><td>0%</td><td>0%</td></tr> <tr> <td>Marine aerosol</td><td>1.1%</td><td>1.0%</td></tr> <tr> <td>Secondary ammonium sulfate</td><td>22%</td><td>23.3%</td></tr> <tr> <td>Secondary ammonium nitrate</td><td>16.2%</td><td>13.3%</td></tr> <tr> <td>Other</td><td></td><td></td></tr> <tr> <td>Non-vehicle OC</td><td>20.6%</td><td>16.0%</td></tr> <tr> <td>Unexplained</td><td>7.4%</td><td>15.2%</td></tr> <tr> <td>Average Measured PM_{2.5} Mass (µg/m³)</td><td>35.6 ± 2.7</td><td>NA</td></tr> <tr> <td>Number in Average</td><td>53</td><td>NA</td></tr> </table> <p>Sihwa site was affected by both local anthropogenic sources and long-range transport of air pollutants.</p> <p>Contributions from incineration were highest when winds were from the southwest or west.</p>	Source Type	Average SCE of 5 intensives	Average over SW and W wind	Industry			Heavy oil combustion	0.3%	0.7%	Incinerator	4.9%	6.1%	Metallurgy	0.8%	0.8%	Copper industry	0.5%	0.7%	Transportation (diesel exhaust)	8.6%	7.2%	Vegetative burning (agricultural burning)	11.3%	10.5%	Geological			Road dust	6.4%	5.0%	Construction	0%	0%	Marine aerosol	1.1%	1.0%	Secondary ammonium sulfate	22%	23.3%	Secondary ammonium nitrate	16.2%	13.3%	Other			Non-vehicle OC	20.6%	16.0%	Unexplained	7.4%	15.2%	Average Measured PM _{2.5} Mass (µg/m ³)	35.6 ± 2.7	NA	Number in Average	53	NA
Source Type	Average SCE of 5 intensives	Average over SW and W wind																																																									
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Number in Average	53	NA																																																									

Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,t,t}	Source Apportionment Method [†]	Findings																																																																															
Reference: Central Taiwan PM _{2.5} and PM ₁₀ study. ^{bt} When: Daily 24-hr sampling during 1 intensive period in 1992 (Nov. 5-7), 4 intensive periods in 1993 (Feb. 8-10, May 13-15, Aug. 10-16, Nov. 12-14), and 1 intensive period in 1993-94 (Dec. 29-Jan. 3). Where: Central Taiwan, at three coastal sites (Dacheng, Meliao, and Taihsi) near agricultural and fishing communities with a coal-fired power plant and many small brick kilns ~50 km north of the study area. A new petrochemical complex is under construction at the village of Meliao. Ambient: PM _{2.5} , PM _{coarse} , and particle size fractions were acquired on Teflon and quartz filters with dichotomous and MOUDI samplers. Samples were analyzed for mass, 20 elements (Na to Pb by XRF), ions (NO ₃ and SO ₄ by IC), and carbon (OC and EC by elemental analyzer, CHN-O RAPID). Source: No area-specific source profile measurements were made. Source profiles applied were reported without associated uncertainties.	Solution: Effective variance weighted least squares CMB7. ^b Source profiles: Industry (coal- and oil-fired brick kilns ^{bu} , heavy oil burning [#13502] ^{ae} , phosphorus plant ^{ac}) Transportation (automobile exhaust [#31203 for auto, #32101 for diesel]) ^{ae} Vegetative burning (rice straw, corn stalks, sugarcane leaves) ^{bv,bw} Geological (soil, paved and unpaved road dust) ^{bv,bw} Marine aerosol (#43101) ^{ae} Secondary sulfate ^{ae} CMB fitting parameters: 23 elements (Na to Pb), ions (NO ₃ , SO ₄), and carbon (OC, EC) from 74 each PM _{2.5} and PM _{coarse} samples were used in CMB attribution to as many as 9 PM _{2.5} source types and 4 PM _{coarse} source types. Sensitivity tests: Reported with 6 trial runs. Goodness of fit: 82% of the samples had performance measures of R ² > 0.9, χ ² < 2, and % mass = 75%-100%. Measurement/modeling uncertainties: Not reported. Validation: None.	Average CMB-calculated source contributions to PM _{2.5} and PM _{coarse} (in % mass): <table><tr><th rowspan="2">Source Type</th><th colspan="2">PM_{2.5}</th><th colspan="2">PM_{coarse}</th></tr><tr><th>Summer Aug. 10-16 '93</th><th>Winter Dec. 29 '93 to Jan. 3-9 '94</th><th>Summer Aug. 10-16 '93</th><th>Winter Dec. 29 '93 to Jan. 3-9 '94</th></tr><tr><td>Industry</td><td></td><td></td><td></td><td></td></tr><tr><td>Coal- and oil-fired brick kilns</td><td>16.4%</td><td>13.1%</td><td>12.2%</td><td>25.2%</td></tr><tr><td>Heavy oil burning</td><td>2.2%</td><td>0%</td><td>0%</td><td>0%</td></tr><tr><td>Phosphorus plant</td><td>5.6%</td><td>1.7%</td><td>0%</td><td>0%</td></tr><tr><td>Transportation</td><td></td><td></td><td></td><td></td></tr><tr><td>Gasoline-fueled vehicles</td><td>1.2%</td><td>1.1%</td><td>0%</td><td>0%</td></tr><tr><td>Diesel-fueled vehicles</td><td>8.6%</td><td>7.5%</td><td>0%</td><td>0%</td></tr><tr><td>Vegetative Burning (agricultural waste)</td><td>33.7%</td><td>39.1%</td><td>38.8%</td><td>34.2%</td></tr><tr><td>Geological (soil and road dust)</td><td>8.9%</td><td>3.8%</td><td>34.9%</td><td>21.1%</td></tr><tr><td>Marine aerosol</td><td>0.2%</td><td>4%</td><td>2.8%</td><td>11.4%</td></tr><tr><td>Secondary sulfate</td><td>18.5%</td><td>18.5%</td><td>0%</td><td>0%</td></tr><tr><td>Unexplained</td><td>4.7%</td><td>11.2%</td><td>11.3%</td><td>8%</td></tr><tr><td>Average Measured PM_{2.5} and PM_{coarse} Mass (μg/m³)</td><td>40.9±12.9</td><td>14.5±5.8</td><td>44.4±20.7</td><td>30.9±11.9</td></tr><tr><td>Number in Average</td><td>22</td><td>17</td><td>22</td><td>17</td></tr></table> Out of 6 study periods, vegetative burning (31%-40%) was the largest contributor to PM _{2.5} and PM _{coarse} . Contributions from fossil fuels (coal- and oil-fired kilns) constituted 11%-14% of PM _{2.5} and 7%-25% of PM _{coarse} and were variable from season to season, with small contributions (0%-2.4%) to PM _{2.5} from heavy oil combustion. Base-year source contributions will be compared with results of future studies when a new petrochemical complex is established in the Meliao area to evaluate its environmental impacts.	Source Type	PM _{2.5}		PM _{coarse}		Summer Aug. 10-16 '93	Winter Dec. 29 '93 to Jan. 3-9 '94	Summer Aug. 10-16 '93	Winter Dec. 29 '93 to Jan. 3-9 '94	Industry					Coal- and oil-fired brick kilns	16.4%	13.1%	12.2%	25.2%	Heavy oil burning	2.2%	0%	0%	0%	Phosphorus plant	5.6%	1.7%	0%	0%	Transportation					Gasoline-fueled vehicles	1.2%	1.1%	0%	0%	Diesel-fueled vehicles	8.6%	7.5%	0%	0%	Vegetative Burning (agricultural waste)	33.7%	39.1%	38.8%	34.2%	Geological (soil and road dust)	8.9%	3.8%	34.9%	21.1%	Marine aerosol	0.2%	4%	2.8%	11.4%	Secondary sulfate	18.5%	18.5%	0%	0%	Unexplained	4.7%	11.2%	11.3%	8%	Average Measured PM _{2.5} and PM _{coarse} Mass (μg/m ³)	40.9±12.9	14.5±5.8	44.4±20.7	30.9±11.9	Number in Average	22	17	22	17
Source Type	PM _{2.5}			PM _{coarse}																																																																													
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Number in Average	22	17	22	17																																																																													

Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																																																												
<p>Reference: Southern Taiwan PM_{2.5} and PM₁₀ study.^{bx}</p> <p>When: Two 12-hr (7am to 7pm) samples collected daily during February/March 1999.</p> <p>Where: Southern Kaohsiung (the second-largest city in Taiwan) at three locations (Tzuoying [north], Chianjen [central], and Shuangang [south]) with coal- and oil-fired power plants, refinery, plastic and steel production, and cement kilns in the area.</p> <p>Ambient: PM_{2.5} and PM_{coarse} quartz filter samples were acquired with dichotomous samplers. Samples were analyzed for mass, 10 elements (by acid digestion and ICP-MS), 8 ions (Na⁺, K⁺, Ca⁺⁺, Mg⁺⁺, NH₄⁺, Cl⁻, NO₃⁻, and SO₄⁻ by IC), and carbon (TC and EC by elemental/integration analyzer, CHNS/O 1180).</p> <p>Source: Source sampling for vehicle exhaust (from tunnel exhaust with automobile and motorcycle fleets), paved road dust, municipal solid waste (MSW) incinerator fly ash, and seawater were acquired via dichotomous sampler and laboratory resuspension. 7 measured source profiles and 2 referenced profiles with associated uncertainties were reported.</p>	<p>Solution: Enrichment factor analysis and correlation matrix were used to identify sources for inclusion to be modeled with effective variance weighted least squares CMB8.^{at}</p> <p>Source profiles:</p> <p>Industry (coal- and oil-fired power plants^{ac}, petroleum refinery^{ac}, steel plant^{ac}, cement plant^{ac}, incinerator^{ac}, fertilizer plant^{ac})</p> <p>Transportation (gasoline vehicle exhaust^{ac}, diesel vehicle exhaust^{ac}, tunnel motorcycle exhaust, tunnel gasoline vehicle exhaust)</p> <p>Vegetative burning (outdoor agricultural burning)^{by}</p> <p>Geological (soil, construction dust, paved road dust, limestone)^{bu}</p> <p>Marine aerosol^{ac}</p> <p>Secondary aerosol (ammonium sulfate, ammonium nitrate)^{ac}</p> <p>CMB fitting parameters: 20 species (10 elements, ions [Na⁺, K⁺, Ca⁺⁺, Mg⁺⁺, Cl⁻, NO₃⁻, SO₄⁻], and carbon[OC, EC]) from 45-48 samples were used in CMB attributions to as many as 9 source types.</p> <p>Sensitivity tests: Not reported.</p> <p>Goodness of fit: Not reported.</p> <p>Measurement/modeling uncertainties: Reported.</p> <p>Validation: None.</p>	<p>Average CMB-calculated source contributions to PM_{2.5} (in % mass):</p> <table><tr><th>Source Type</th><th>Tzuoying (N)</th><th>Chianjen (C)</th><th>Shuangang (S)</th></tr><tr><td>Industry</td><td></td><td></td><td></td></tr><tr><td>Power plant</td><td>2%</td><td>0%</td><td>1%</td></tr><tr><td>Petroleum refinery</td><td>2%</td><td>3%</td><td>0%</td></tr><tr><td>Cement kiln</td><td>2%</td><td>0%</td><td>0%</td></tr><tr><td>Transportation</td><td>45%</td><td>18%</td><td>54%</td></tr><tr><td>Vegetative burning (outdoor agricultural burning)</td><td>13%</td><td>17%</td><td>17%</td></tr><tr><td>Geological</td><td></td><td></td><td></td></tr><tr><td>Paved road dust</td><td>2%</td><td>17%</td><td>1%</td></tr><tr><td>Limestone</td><td>0%</td><td>0%</td><td>1%</td></tr><tr><td>Marine aerosol</td><td>4%</td><td>4%</td><td>0%</td></tr><tr><td>Secondary ammonium sulfate</td><td>14%</td><td>22%</td><td>15%</td></tr><tr><td>Secondary ammonium nitrate</td><td>16%</td><td>19%</td><td>12%</td></tr><tr><td>Average Measured PM_{2.5} Mass (μg/m³)</td><td>48.2</td><td>42.7</td><td>53.7</td></tr><tr><td>Number in Average</td><td>15-16</td><td>15-16</td><td>15-16</td></tr></table> <p>Major contributors to PM_{2.5} are motor vehicle exhaust (18%-54%), secondary aerosol (30%-41%), and vegetative burning (13%-17%). Contributions from industries with fossil fuel are low, ranging from 1% to 6% of PM_{2.5} mass.</p> <p>Long-range transport of SO₂ and sulfur oxides from China during winter/spring with prevailing northerly winds may have contributed to elevated secondary aerosol.</p> <p>Both U.S. EPA and local profiles for road dust and vehicle exhaust were assembled, and it is not clear which profiles were used in CMB calculations.</p>	Source Type	Tzuoying (N)	Chianjen (C)	Shuangang (S)	Industry				Power plant	2%	0%	1%	Petroleum refinery	2%	3%	0%	Cement kiln	2%	0%	0%	Transportation	45%	18%	54%	Vegetative burning (outdoor agricultural burning)	13%	17%	17%	Geological				Paved road dust	2%	17%	1%	Limestone	0%	0%	1%	Marine aerosol	4%	4%	0%	Secondary ammonium sulfate	14%	22%	15%	Secondary ammonium nitrate	16%	19%	12%	Average Measured PM _{2.5} Mass (μg/m ³)	48.2	42.7	53.7	Number in Average	15-16	15-16	15-16
Source Type	Tzuoying (N)	Chianjen (C)	Shuangang (S)																																																											
Industry																																																														
Power plant	2%	0%	1%																																																											
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Number in Average	15-16	15-16	15-16																																																											

Table 1. (Continued)

Study, Location, Period, and Measurements ^{a,†,‡}	Source Apportionment Method [‡]	Findings																											
<p>Reference: McMurdo Station, Antarctica, PM₁₀ study.^{bz,ca,cb}</p> <p>When: Continuous 48-hr sampling from Nov. 1995 through Jan. 1996 and from Nov. 1996 through Jan. 1997 (austral summers).</p> <p>Where: McMurdo Station, Ross Island, Antarctica, at 2 locations. The Hut Point site is 1 km north-west and downwind of McMurdo, and the Radar Sat site is on a ridge 2.5 km north of McMurdo.</p> <p>Ambient: Teflon and quartz PM₁₀ samples and SO₂ (on impregnated Whatman 41 cellulose-fiber filters) samples were collected with SFS and hivol SSI samplers. SFS samples were analyzed for PM₁₀ mass, 38 elements (Al to Pb by XRF), ions (Cl⁻, NO₃⁻, SO₄²⁻ and MSA by IC; NH₄⁺ by AC; and water-soluble Na⁺ and K⁺ by AAS), carbon (OC and EC by TOR), and SO₂ (as SO₄²⁻ by IC). Hivol samples were analyzed for elements (by INAA).</p> <p>Source: Source profiles from 4 diesel-fueled power generators, 5 space heaters, 5 steam-heated baths, surface light-duty gasoline vehicle exhaust, high and low idles of helicopters, 8 C-130 aircraft exhaust, and 10 local soil samples were used. Source profiles with associated uncertainties are reported in Mazzera et al.^{bz}</p>	<p>Solution: Effective variance weighted least squares CMB7.^b</p> <p>Source profiles:</p> <ul style="list-style-type: none"> Industry (diesel-fueled power plant, heating exhaust) Transportation (gasoline vehicles, diesel vehicles, diesel equipment, helicopters, C-130 aircraft) Secondary aerosol (ammonium sulfate, ammonium nitrate)^g <p>CMB fitting parameters: 21 elements (Al to Pb), ions (NO₃⁻, SO₄²⁻, NH₄⁺, Na⁺, K⁺), and carbon (OC, EC) were used in CMB attributions to 7 source types.</p> <p>Sensitivity tests: Reported in Mazzera et al.^{bz}</p> <p>Goodness of fits: Reported in Mazzera et al.^{bz}</p> <p>Measurement/modeling uncertainties: Reported.</p> <p>Validation: The relationship between NSS, excess sulfate (sulfate not associate with geological dust, sea salt, or primary combustion emissions), and MSA were used to apportion sulfate.</p>	<p>Average CMB-calculated source contributions to PM₁₀ and sulfate at the Hut Point site (in % mass)</p> <table> <tr> <th>Source Type</th><th>PM₁₀</th><th>Aerosol sulfate</th></tr> <tr> <td>Industry and transportation (diesel-power generation and vehicle exhaust)</td><td>14%</td><td>20%</td></tr> <tr> <td>Geological</td><td>57%</td><td>12%</td></tr> <tr> <td>Marine aerosol</td><td>15%</td><td>8%</td></tr> <tr> <td>Other (biogenic sulfur)</td><td>3%</td><td>—</td></tr> <tr> <td>Secondary ammonium sulfate</td><td>10%</td><td>60%[†]</td></tr> <tr> <td>Secondary ammonium nitrate</td><td>1%</td><td>—</td></tr> <tr> <td>Average measured PM₁₀ mass (µg/m³)</td><td>3.4 ± 0.2</td><td>0.46 ± 0.03</td></tr> <tr> <td>Number in Average</td><td>50</td><td>50</td></tr> </table> <p>† 36% attributed to biogenic sources and 24% to other sources.</p> <p>69% of contributions from diesel-powered generators was attributed to the McMurdo power generating station for electricity and water production, and 31% to space heating.</p> <p>Mt. Erebus may be a distant sulfur source in the Ross Island area.</p>	Source Type	PM ₁₀	Aerosol sulfate	Industry and transportation (diesel-power generation and vehicle exhaust)	14%	20%	Geological	57%	12%	Marine aerosol	15%	8%	Other (biogenic sulfur)	3%	—	Secondary ammonium sulfate	10%	60% [†]	Secondary ammonium nitrate	1%	—	Average measured PM ₁₀ mass (µg/m ³)	3.4 ± 0.2	0.46 ± 0.03	Number in Average	50	50
Source Type	PM ₁₀	Aerosol sulfate																											
Industry and transportation (diesel-power generation and vehicle exhaust)	14%	20%																											
Geological	57%	12%																											
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Secondary ammonium sulfate	10%	60% [†]																											
Secondary ammonium nitrate	1%	—																											
Average measured PM ₁₀ mass (µg/m ³)	3.4 ± 0.2	0.46 ± 0.03																											
Number in Average	50	50																											

Table 1. (Continued)

* Monitoring instruments (see Hering^{cc} and Watson and Chow^{cd} for descriptions and more detailed references):

Dichotomous	dichotomous sampler (Andersen Instruments, Smyrna, GA)
DSP	dilution source probe ^{cc}
Hivol SSL	high-volume sampler with PM ₁₀ size-selective inlet (Andersen Instruments, Smyrna, GA)
IMPROVE	Interagency Monitoring of Protected Visual Environments sampler, used in the IMPROVE visibility network in the U.S. and in the Guelph atmospheric visibility monitoring network in Canada (University of California, Davis, CA)
Minivol	Minivol sampler (PM ₁₀ , PM _{2.5}) (Airmetrics, Eugene, OR)
MOI	micro-orifice impactor
MOUDI	Micro-Orifice Uniform Deposit Impactor sampler (MSP, Minneapolis, MN)
Partisol	Partisol Model 2300 sequential speciation sampler (PM ₁₀ , PM _{2.5}) (Rupprecht & Patashnick, Albany, NY)
SCAQs	Southern California Air Quality Study sampler (California Air Resources Board, Sacramento, CA)
SFS	medium-volume sequential filter sampler (Desert Research Institute, Reno, NV)
SGS	medium-volume sequential gas sampler (Desert Research Institute, Reno, NV)
URG	URG cyclone sampler (URG, Chapel Hill, NC)
VAPS	Versatile Air Pollutant Sampler (URG, Chapel Hill, NC)

† Chemical measurements (see Chow^{cf} for descriptions and more detailed references):

AAS	atomic absorption spectrometry for water-soluble Na ⁺ and K ⁺
AC	automated colorimetry for NH ₄ ⁺
CHN-O RAPID	elemental analyzer with integration analyzer (W.C. Heraeus, Germany)
CHNS/O 1180	elemental analyzer with DP700 integration analyzer ^{bx} (CE Instruments, Milan, Italy)
GC-MS	gas chromatography with mass spectrometry for n-alkanes, organic acids, and PAHs
HRGC	high-resolution gas chromatography
IC	ion chromatography for Cl ⁻ , NO ₃ ⁻ , SO ₄ ²⁻ , NH ₄ ⁺ , Na ⁺ , and/or K ⁺
IC/CE	ion chromatography with capillary electrophoresis
ICP/MS	inductively coupled plasma with mass spectrometry analysis for elements
INAA	instrumental neutron activation analysis for elements
LIPM	laser integrating plate method for particle light absorption
PESA	proton elastic scattering analysis for organic carbon estimates from hydrogen abundance developed by the University of California, Davis, for the IMPROVE network
PIXE	proton-induced x-ray emission spectroscopy for elements
SEM	scanning electron microscopy for particle morphology
SEM/EDX	scanning electron microscopy with energy dispersive x-ray fluorescence analysis for particle morphology
TMO	thermal manganese oxidation for OC and EC ^{cg}
TOA	thermal/optical analysis for OC and EC
TOR	thermal/optical reflectance for OC and EC ^{ch,ci}
TOT	thermal/optical transmission for OC and EC ^{cj}
XRF	x-ray fluorescence spectroscopy for elements ^{ck}

Table 1. (Continued)

† Mass and chemical species:

Al.....	aluminum	NH ₄ ⁺	ammonium
As.....	arsenic	NH ₄ NO ₃	secondary ammonium nitrate
B(a)P.....	benzo[a]pyrene	(NH ₄) ₂ SO ₄	secondary ammonium sulfate
b _{abs}	particle light absorption	NH ₄ HSO ₄	secondary ammonium bisulfate
b _{ext}	gas and particle light extinction	Ni.....	nickel
Br.....	bromine	NO ₃ ⁻	nitrate
Ca ⁺⁺	water-soluble calcium	NSS.....	non-sea-salt sulfate, total sulfate minus (0.25 × sodium)
Cl ⁻	chloride	OC.....	organic carbon
Cr.....	chromium	PAH.....	polycyclic aromatic hydrocarbons
Cu.....	copper	Pb.....	lead
EC.....	elemental carbon	PM ₁₀	particulate matter with aerodynamic diameters less than 10 μm
F ⁻	fluoride	PM _{2.5}	particulate matter with aerodynamic diameters less than 2.5 μm
Fe.....	iron	PM _{coarse}	particulate matter with aerodynamic diameters between 2.5 and 10 μm
HC.....	hydrocarbons	S.....	sulfur
HCl.....	hydrogen chloride	SAS.....	spherical aluminosilicate particles
HF.....	hydrogen fluoride	Se.....	selenium
HNO ₃	gaseous nitric acid	Si.....	silicon
K.....	potassium	SO ₂	sulfur dioxide
K ⁺	water-soluble potassium	SO ₄ ²⁻	sulfate
Mo.....	molybdenum	SO _x	total sulfur oxides (sum of SO ₂ and particulate SO ₄)
Mg ⁺⁺	water-soluble magnesium	TC.....	total carbon
MSA.....	methane sulfonate	VOCs.....	volatile organic compounds
Na ⁺	water-soluble sodium	Zr.....	zirconium
NH ₃	gaseous ammonia		

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(l)	117	(ab)	50	(au)	143
(m)	South Coast Air Quality Management District. "Emissions inventory South Coast Air Basin, Working Paper No. 1, 1987 AQMP Revision". Prepared by South Coast Air Quality Management District: Diamond Bar, CA, 1986.	(ac)	111	(av)	55
(n)	Core, J. E.; Houck, J. E. "Pacific Northwest Source Profile Library Sampling and Analytical Protocols". Prepared by Oregon Department of Environmental Quality: Portland, OR, 1987.	(ad)	Pytkowicz, R. M.; Kester, D. R. <i>Oceanogr. Mar. Biol.</i> , 1971 , 9, 11-60.	(aw)	74
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(t)	132	(aj)	Gray, H. A.; Cass, G. R.; Huntzicker, J. J.; Heyerdahl, E. K.; Rau, J. A. <i>Environ. Sci. Technol.</i> , 1986 , 20(6), 580-589.	(bc)	101
(u)	126	(ak)	Gray, H. A. Control of atmospheric fine primary carbon particle concentrations. Ph.D. Thesis, California Institute of Technology, Pasadena, CA, 1986.	(bd)	57
(v)	131	(al)	52		
(w)	Hildemann, L. M.; Markowski, G. R.; Cass, G. R. <i>Environ. Sci. Technol.</i> , 1991 , 25(4), 744-759.	(am)	78		
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(z)	137	(ap)	75		
		(aq)	54		
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Grand Canyon^{56,57} and one determined the sources of PM_{2.5} causing haze near the Mt. Zirkel Wilderness in northwestern Colorado.⁵⁸ Studies outside the United States included the vicinity of Vancouver, British Columbia,⁵⁹ and Toronto, Ontario,⁶⁰ in Canada; Mexico City, Mexico;⁶¹ an industrial area northwest of Prague in the Czech Republic;⁶² a township south of Johannesburg, South Africa;⁶³ south of Seoul in Korea;^{64,65} near Taichung⁶⁶ and southern Kaohsiung⁶⁷ in Taiwan; and at McMurdo Station in the Antarctic.⁶⁸

CMB Study Goals and Objectives. Most of these studies were undertaken to support decision-making. PM₁₀ source apportionments for Los Angeles,⁵¹ California's Imperial Valley,⁵² and Las Vegas⁵³ were used to develop State Implementation Plans to attain the PM₁₀ NAAQS. Fugitive dust and/or ammonia (NH₃) reduction rules have been promulgated in these areas. The Denver PM_{2.5} study⁵⁴ was completed, in part, to evaluate the success of wood burning controls implemented since a prior study in 1987–88,⁶⁹ and major improvements were demonstrated. The Grand Canyon^{56,57} and Mt. Zirkel⁷⁰ studies provided information about the effects of single

emissions sources on perceptible visibility impairment. Sulfur dioxide scrubbers are to be installed on coal-fired power stations in both areas as a result of these studies and other considerations. Evaluating the effects of addition or removal of an industrial facility were the objects of the Chicago study,⁵⁵ in which monitoring was conducted before and after operation of a Waste to Energy power generator, and for Mexico City,⁶¹ in which measurements were taken prior to closure of an old oil refinery for later comparison with source contribution estimates after operations ceased. The CMB was used in South Africa⁶³ to evaluate the effects of different "low-smoke" coals on air quality in townships that cook and heat with these domestic fuels.

CMB source apportionment studies in San Jose, CA,⁴⁷ San Joaquin Valley, CA,^{48,71} Santa Barbara, CA,⁵⁰ Korea,⁶⁴ Taiwan,^{66,72} and Antarctica⁶⁸ were informational rather than regulatory; there was a desire by decision-makers to understand the relative contributions from different source types. The information is also useful to inform people within a jurisdiction of the major and minor contributors to PM_{2.5} and PM₁₀ when complaints arise. Studies in the San Joaquin Valley, CA,⁴⁹ Los Angeles, CA,⁴⁵ Denver, CO,⁵⁴ and Czech Republic⁶² had research components that explored how to use more specific organic compounds measured in source and receptor samples to distinguish among a larger number of source categories.

Particle Size Fractions. Seven of the studies estimated source contributions to PM₁₀; eleven estimated contributions to PM_{2.5}; one to both PM_{2.5} and PM_{coarse} (PM₁₀ minus PM_{2.5}) fractions;⁶⁶ one to PM_{2.5} carbon, SO₂, and NO_x;⁵⁴ one to trace elements;⁵⁶ and one to sulfate and SO_x (SO₂ plus sulfate).⁵⁷ PM₁₀ studies were primarily motivated by the existing PM₁₀ NAAQS⁷³ while PM_{2.5} studies were motivated by the anticipation of a PM_{2.5} NAAQS, poor visibility, or a focus on industrial and mobile rather than fugitive dust emissions. PM_{coarse} is typically dominated by fugitive dust contributions, whereas PM_{2.5} contains most of the primary and secondary contributions from fossil and biogenic fuel combustion.

Ambient Measurements. All of the CMB applications used chemical measurements of elements (a subset of 40 elements from Na to U), water-soluble ions (chloride [Cl⁻], nitrate [NO₃⁻], sulfate [SO₄²⁻], ammonium [NH₄⁺], and sometimes sodium [Na⁺], potassium [K⁺], calcium [Ca²⁺]), and carbon (organic [OC]

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and elemental carbon [EC] or black carbon [BC]). Specific organic compounds (such as polycyclic aromatic hydrocarbons [PAH], *n*-alkanes, *n*-alkenes, organic acids, hopanes, sterenes, lactones, sterols, guaiacols, and syringols) were measured in several cases. Cooking and wood burning source contributions were shown to be distinguishable using several of these compounds,^{45,49,54} implying that the vegetative burning category determined without them should be redefined as a cooking/vegetative burning category. It was possible to divide the motor vehicle exhaust category into diesel and gasoline engine fractions using these markers.^{20,45,49} Microscopic properties such as aluminosilicate cenospheres were used explicitly as a CMB fitting species⁵⁷ and as a qualitative verification of contributions from coal-combustion.⁶²

Source Measurements and Profiles. Several new source profiles were measured for the areas and times of these studies, including diesel-powered electrical generation,⁶⁸ coal-fired power plant fly ash and stack emissions,^{55,62,63,74} refuse-derived fuel electrical generator fly ash,⁵⁵ manure-fired electrical generator stack emissions,^{20,76} gasoline and diesel vehicle exhaust,^{20,62,63,72,74–76} paved and unpaved road dust,^{20,47,50,63,74–78} disturbed soil,⁷⁵ residential coal stoves and heaters,^{62,63,74} wood stoves and fireplaces,^{20,74,75} agricultural burning,^{63,74,76} meat cooking,^{54,76} lime kiln,⁶³ incinerator,^{62,72} oil-fueled glass plant,⁷⁶ steel/metal production,⁶³ and geothermal springs.⁷⁴ Other profiles were taken from earlier tests in the study area or similar areas. Some profiles were obtained from the U.S. EPA's SPECIATE⁷⁹ database.

Because source profiles are measured at the source, they represent the composition of emissions before changes have taken place owing to atmospheric transformation and deposition. Much of the sulfate, nitrate, and ammonium is formed en route between source and receptor and is not represented by the primary emissions profile. To account for this, most of the CMB applications included profiles for secondary inorganic aerosols, typically some combination of sulfuric acid (H_2SO_4), ammonium bisulfate (NH_4HSO_4), ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$), ammonium nitrate (NH_4NO_3), and sodium nitrate (NaNO_3). These were represented as stoichiometric weight ratios for the potential secondary end-products (e.g., 27% NH_4 and 73% SO_4 for $(\text{NH}_4)_2(\text{SO}_4)$). Most of the studies found that the ammonium sulfate and ammonium nitrate profiles best explained the ambient measurements, indicating a regional mix

of SO_2 and NO_x emissions that converted to sulfuric and nitric acids and had ample opportunity to encounter neutralizing NH_3 from biogenic and other emitters. Mazzera et al.⁶⁸ found that a NH_4HSO_4 profile best fit the ambient measurements, consistent with more local formation that used up available NH_3 , as opposed to a regional mixture. Chow et al.⁵⁰ used profiles that assumed 0%, 25%, 50%, 75%, and 100% reaction of nitric acid with the sodium chloride in a marine profile, as described by Watson et al.,⁸⁰ to determine the degree of interaction between urban (nitric acid) and natural (sea salt) contributions.

Watson et al.⁵⁸ used an aerosol evolution model, as suggested by Lewis and Stevens⁸¹ and Gordon and Olmez,⁸² to create “aged” profiles that accounted for deposition and transformation of chemical components in primary particles, SO_2 , and SO_4^{2-} formed from gas as a function of aging for wet and dry conditions. Using SO_2 , SO_4^{2-} , and the stack emission elements in the CMB, they estimated an upper limit on $\text{PM}_{2.5}$ and sulfate contributions from local power stations distinct from the regional ammonium sulfate.

Eatough et al.⁵⁷ estimated regional profiles on the basis of measurements in source areas from periods that were meteorologically isolated from contamination by other source areas, similar to the method first used in the eastern United States by Rahn and Lowenthal.⁸³ These profiles were similar, and indistinguishable by the CMB, when they just contained urban emissions, but they did show differences when elemental and morphological markers for smelters and coal-fired power generation were located in the source region.

Several of the studies estimated a “secondary organic carbon” contribution by assuming that organic carbon not assigned to another source resulted from the conversion of heavy organic gases to particles.⁸⁴ A “single constituent source type” profile⁸⁵ that contained a 100% abundance for OC and a 0% abundance for all other species was used to estimate the secondary organic carbon.

Source Contribution Estimates. The largest fossil fuel contribution to primary $\text{PM}_{2.5}$ and PM_{10} was attributed to gasoline- and diesel-vehicle exhaust rather than industrial sources in most of these studies. Transportation sources supplied large amounts of OC and EC and small amounts of trace elements such as zinc and iron; they also supplied lead and bromine when leaded gasoline was used.^{45,49,54} Only those studies with organic compounds in ambient measurements and source profiles were able to separate the vehicle exhaust category into gasoline and diesel portions using the CMB. The

(74) Watson, J. G.; Chow, J. C.; Houck, J. E. *Chemosphere* **2001**, *43* (8), 1141–1151.

(75) Chow, J. C.; Watson, J. G. Fugitive dust and other source contributions to PM_{10} in Nevada's Las Vegas Valley; Report No. 4039.1F. Prepared for Clark County Department of Comprehensive Planning, Las Vegas, NV, by Desert Research Institute: Reno, NV, 1997.

(76) Watson, J. G.; Chow, J. C. *Sci. Total Environ.* **2001**, *276* (1–3), 33–47.

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(78) Chow, J. C.; Watson, J. G. Imperial Valley/Mexicali Cross Border PM_{10} Transport Study; Report No. 4692.1D1. Prepared for U.S. Environmental Protection Agency, Region IX, San Francisco, CA, by Desert Research Institute: Reno, NV, 1997.

(79) U.S. EPA. “SPECIATE: EPA's repository of total organic compound and particulate matter speciated profiles for a variety of sources for use in source apportionment studies”. U.S. Environmental Protection Agency OAQPS, Research Triangle Park, NC, 1999. <http://www.epa.gov/ttn/chief/software/speciate>.

(80) Watson, J. G.; Chow, J. C.; Lurmann, F. W.; Musarra, S. *JAWMA* **1994**, *44* (4), 405–412.

(81) Lewis, C. W.; Stevens, R. K. *Atmos. Environ.* **1985**, *19* (6), 917–924.

(82) Gordon, G. E.; Olmez, I. Hybrid receptor modeling with multiple sources and vertical mixing. In *Transactions, Receptor Methods for Source Apportionment: Real World Issues and Applications*; Pace, T. G., Ed.; Air Pollution Control Association: Pittsburgh, PA, 1986; pp 229–238.

(83) Rahn, K. A.; Lowenthal, D. H. *Science* **1984**, *223*, 132–139.

(84) Pandis, S. N.; Harley, R. A.; Cass, G. R.; Seinfeld, J. H. *Atmos. Environ.* **1992**, *26A* (13), 2269–2282.

(85) Watson, J. G. Chemical element balance receptor model methodology for assessing the sources of fine and total suspended particulate matter in Portland, Oregon. Ph.D. Dissertation, Oregon Graduate Center, Beaverton, OR, 1979.

diesel exhaust contribution was estimated to be three to five times the gasoline exhaust contribution for annual average $PM_{2.5}$ in Los Angeles during 1982⁴⁵ and two to five times the gasoline exhaust contribution in Fresno and Bakersfield during winter of 1995–96.⁴⁹ Diesel exhaust contributions were also approximately three times gasoline exhaust contributions for hot stabilized gasoline emissions in Denver, CO, during winter 1996–97, but when profiles for cold start and high-emitting gasoline vehicles were included, the ratio of diesel to gasoline contributions reversed and the ambient concentrations of OC, EC, and organic compounds could be better explained.⁵⁴

The Czech Republic⁶² and South Africa⁶³ neighborhoods where coal was used for home heating, and where economic conditions mitigated against widespread vehicle use, showed residential cooking and heating with raw coal as the largest fossil fuel contributor to primary $PM_{2.5}$. Coal- and oil-fired brick kilns were the largest fossil-fuel contributors in central Taiwan.⁶⁶ An uncontrolled coal-fired power station using high-ash coal in the Czech Republic⁶² and an uncontrolled petroleum refinery in Mexico City⁶¹ were large fossil fuel contributors prior to their modification or closure. With the exception of these uncontrolled emitters^{61,62,66} during the study periods, primary particle contributions to $PM_{2.5}$ and PM_{10} from coal, oil, and natural gas combustion in power stations and industrial facilities were negligible to minor contributors to primary $PM_{2.5}$ and PM_{10} , usually <2% of mass. Their contributions to secondary inorganic sulfates and nitrates is probably higher. The Grand Canyon⁵⁷ and Mt. Zirkel⁷⁰ visibility studies attributed nearly all of the secondary sulfate to coal-fired power stations, primarily on the basis of emissions inventories for the study regions. However, source contributions from specific power stations to this regional sulfate were associated with large standard errors.

Nonfossil fuel contributions included fugitive dust, vegetative burning from residential heating with wood, prescribed burning, or wildfires, other industrial sources, and natural sources such as marine aerosol. Specific organic compounds were used to distinguish emissions from vegetative burning and cooking, with approximately equal amounts of each occurring in samples from Denver, CO,⁵⁴ and Los Angeles, CA.⁴⁵ Very large contributions of wood burning and cooking were found in Fresno, CA, over a period that included the days after Christmas, with much lower contributions found at the same location a week later.⁴⁹ Marine aerosol was a large contributor in coastal areas such as California's central coast⁵⁰ and Antarctica⁶⁸ where manmade contributions were low. Smelters, incinerators, steel making and other industrial sources with emissions controls were negligible (<1%) contributors and typically with high uncertainties for the source contribution estimates. This uncertainty was too large to distinguish between contributions from a Waste-To-Energy (WTE) electrical generator using Refuse Derived Fuel (RDF) in Robbins, IL, for periods before and after it started operating.⁵⁵ The CMB source apportionment did show that the WTE contribution was negligible and undetectable in excess of other sources with similar chemical profiles in this industrial area.

Fugitive dust from paved and unpaved roads and disturbed surfaces as found at construction sites was the largest contributor to most PM_{10} measurements^{53,75} and typically 10% to 20% of $PM_{2.5}$. Fugitive dust contained many of the same elements found in coal stack emissions and fly ash, but usually in different proportions, thereby allowing it to be separated by the CMB. Coal stack emission profiles often included small but detectable abundances of selenium, lead, and arsenic that were depleted in geological material.

Reconciliation with Source Models and Emissions Inventories. Discrepancies between source contributions determined from emissions inventories and those found by receptor models are often due to inappropriate emissions factors, insufficient information about the activities that generate emissions, and emissions sources that have not been included in the inventory. Relative source contribution estimates were compared with results from emissions inventories for the San Joaquin Valley, CA,⁴⁸ Los Angeles, CA,⁴⁵ Las Vegas, NV,⁵³ and Denver, CO.⁵⁴ Figure 1 shows an example from Las Vegas, NV, which was the only study reporting both a source model and an inventory comparison. In this case, the comparison between relative contributions from CMB and Industrial Source Complex (ISC) dispersion model results is good for the E. Charleston site, thereby lending confidence to both the emissions inventory and the source apportionment, at least with respect to the annual average. A poorer comparison was found at the Bemis site, especially for motor vehicle exhaust, indicating that the inventory around the sampler may need improvement. Similarly good comparisons were found between ratios of specific chemical components for CMB source contributions and emissions inventories in the Los Angeles area.⁴⁵ In contrast, $PM_{2.5}$ fugitive dust emissions in Denver, CO,⁵⁴ exceeded those determined by CMB by more than a factor of 2; when cold start and high-emitting vehicle emissions were apportioned by the CMB, the ratio of gasoline to diesel contributions was 4:1, disagreeing with the 1:4 ratio in the wintertime Denver emissions inventory. Cold starts and high emitters were not included in the inventory and accounted for most of the difference in the CMB source contribution estimates.

Discussion

Study Goals and Objectives. Although the CMB was originally developed for quantifying contributions from primary airborne particulate emitters to justify PM_{10} state implementation plans, the studies reviewed here show that it has been adapted and applied for other constructive purposes. The apportionment of secondary aerosols is especially important for fossil fuel combustion as this is the main source of the SO_2 and NO_x precursors. Using aerosol evolution models to estimate how profiles change offers the most promise for secondary aerosol apportionment with the CMB, but the applications reviewed here are fairly primitive. Kleeman and Cass⁸⁶ demonstrate a practical and more precise method of simulating source composition and size changes during transport with interactions among

(86) Kleeman, M. J.; Cass, G. R. *Atmos. Environ.* **1999**, 33 (28), 4597–4613.

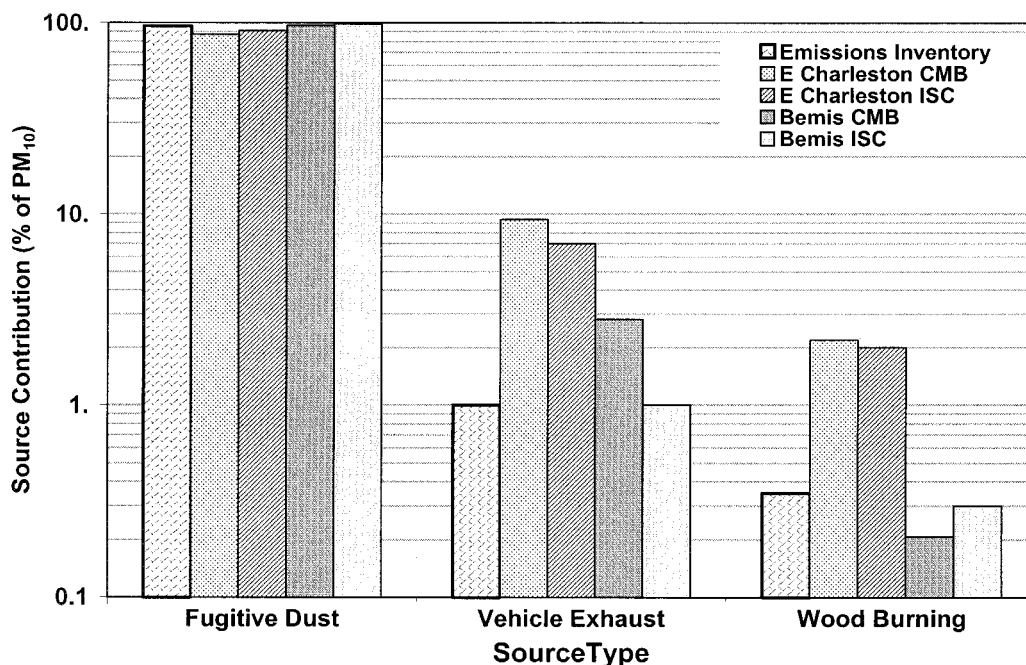


Figure 1. Comparison of relative annual average primary PM₁₀ source contribution estimates at two receptor sites in Las Vegas, NV,⁵³ using the annual emissions inventory, the CMB receptor model, and the Industrial Source Complex (ISC) dispersion model. The Bemis site was in an industrial area on the north side of the city surrounded by construction activities during the 1995 monitoring period. The E. Charleston site was in an established neighborhood without nearby construction in the vicinity of heavily traveled roadways. Secondary sulfate and secondary nitrate source contributions were subtracted from the measured PM₁₀ for comparison with the primary particle inventory.

emissions from several sources. This type of source model could create aged profiles for selected receptors and meteorological regimes over a short time period during which it is practical to obtain the detailed emissions, meteorological, and chemical measurements the source model requires as input. These aged profiles could then be used for several years thereafter to estimate secondary source contributions by CMB with only the receptor measurements required.

An important new objective is the verification and improvement of emissions inventories. Although rates for ducted pollutants at industrial facilities can be accurately quantified, primary particle and precursor emissions from area sources such as heating, cooking, vehicle exhaust, fugitive dust, and NH₃ creators are highly uncertain. This uncertainty derives from inaccurate activity estimates (i.e., number of vehicle miles traveled, amount of wood burned) as well as nonrepresentative emissions factors. It also derives from imperfect spatial and temporal allocation of emission rates to areas smaller than a city, county, or state, and to periods of the day, days of the week, and seasons of the year.

CMB applications are often criticized because an insufficient number of sources⁷⁹ have been sampled to adequately represent their chemical composition in a study region. The critics ignore or are unaware of the fact that emissions factors are derived from even fewer tests that practically *never* correspond to the time period or region being studied. For example, Lloyd and Cackette⁸ reveal that all diesel engine emissions in California are based on only 70 source tests over a 30-year period. Emissions factors have no upper limits, whereas the abundances within a source profile are constrained to values between 0% and 100% and the sum of all abundances must be less than or equal to

100%. Emissions factors have no independent constraints, whereas the combination of source profiles used in the CMB must reasonably reproduce the PM_{2.5} and PM₁₀ mass (within $\pm 20\%$) and the receptor chemical measurements (within two standard deviations of the ambient data and source contribution estimate uncertainty intervals). More importantly, emissions inventories are often reported to three or four significant digits with no precision estimates, whereas every effective variance CMB source contribution estimate is accompanied by a standard error that is a function of the input data uncertainties and the collinearity of source profiles.

There will never be sufficient resources for enough tests of source profiles or emissions factors to know that the available tests represent a specific situation. An independent reconciliation with ambient data using a CMB apportionment, similar to that illustrated in Figure 1, can focus available resources on portions of the inventory that have the largest discrepancies. Fugitive dust often constitutes 80% to 90% of primary emissions in PM₁₀ inventories, but only ~50% of PM₁₀ measured in the air. This is an important emissions inventory category that needs to be reexamined.^{87,88} Cooking is an important PM_{2.5} contributor according to the CMB, but it is not even considered in most inventories. Lacking specific organic compounds such as cholesterol for cooking and levoglucosan, guaiacols, and

(87) Watson, J. G.; Chow, J. C.; Pace, T. G. Fugitive dust emissions. In *Air Pollution Engineering Manual*, 2nd ed.; Davis, W. T., Ed.; John Wiley & Sons, Inc.: New York, 2000; pp 117–135.

(88) Countess, R. J.; Barnard, W. R.; Claiborn, C. S.; Gillette, D. A.; Latimer, D. A.; Pace, T. G.; Watson, J. G. Methodology for estimating fugitive windblown and mechanically resuspended road dust emissions applicable for regional scale air quality modeling; Report No. 30203-9. Prepared by Western Regional Air Partnership; Denver, CO, 2001.

syringols for vegetative burning, cooking contributions have usually been classified with wood and agricultural burning and thereby exempted from pollution controls. High emitters and cold starts from gasoline vehicles were determined to be important contributors in wintertime Denver, CO, but the situation may be different in other seasons and other climates. Mobile source inventories in Colorado and elsewhere need to be reexamined in light of these CMB findings.

Ambient Measurements. Elements, ions, and carbon fractions are essential to obtaining a mass balance and apportioning major source categories such as coal-fired power plants, residual oil burning, smelters, refineries, steel-making, residential solid waste combustion, gasoline/diesel exhaust, cooking/vegetative burning, fugitive dust, marine aerosol, and secondary sulfates, nitrates, and organics.

Many receptor modeling studies from the 1970s and 1980s used only elemental concentrations, and large portions of the mass could not be accounted for without carbon, nitrate, and ammonium measurements. Secondary, water-soluble sulfate was estimated as three times the sulfur concentrations, and this ignored nonsulfate sulfur and insoluble primary emissions such as organic sulfates, SO₂ adsorbed onto soot (BC) particles, gypsum, and fugitive dust from solid sulfur handling.^{89,90} Earlier studies did quantify several industrial contributions to trace metals, many of which were believed to be toxic, and efforts were made to reduce their emissions. These emissions reductions were successful, but they removed some of the common indicators for these sources. Nickel (Ni) and vanadium (V) were good indicators of heavy residual oil combustion, but this fuel is rarely used for power generation and heating in cities in the United States today. In addition to air quality considerations, many refineries installed fluidized catalyst crackers in the 1980s that allowed much of the heavy oil to be converted to higher value products. Small amounts of V and Ni are released when the catalyst is cleaned, but typically after passing through efficient particle removal systems. Although regulated in harbors, many ships operating in international waters off the Pacific and Atlantic coasts of the United States and elsewhere^{91,92} still use heavy and waste oils with high sulfur and metal contents. Mazzera et al.⁶⁸ found the McMurdo Station supply ship to dominate the PM₁₀ and SO₂ concentrations when it docked near the Hut Point monitor.

Lead (Pb) and bromine (Br) were the main indicators of gasoline fuel use, owing to the addition of tetraethyl lead. Vega et al.⁶¹ took advantage of this to quantify vehicle emissions for 1990 in Mexico City, but Pb has been phased out of Mexico gasoline today and Pb and Br are no longer useful markers distinguishing between gasoline and diesel exhaust. Manganese (Mn) was once

added to gasoline as an octane enhancer,⁹³⁻⁹⁷ but human toxicity considerations have mitigated against this use in the United States. Barium (Ba) was once used as a smoke inhibitor for diesel fuel,⁹⁸ but this was discontinued for the same health reasons.

Nonducted gasoline/diesel exhaust (OC, EC), vegetative burning/cooking (OC, EC, K⁺), fugitive dust (Al, Si, Ti, Ca), and marine/playa/de-icing (Na⁺, Cl⁻) source type contributions can be quantified by the CMB, with the parenthetical components being most influential for each source type. Primary contributions from ducted industrial sources containing elements uncommon to crustal material can also be quantified, such as refinery/residual oil (Ni, V), steel making (Fe, Mn, Pb), coal burning (Se, As), smelting (Cu, Zn, Pb), and cement manufacturing (Ca). The quantity and chemical form of secondary sulfates and nitrates can also be determined. As shown in this classification, the source types must be broadly defined to include the subtypes that cannot be distinguished by receptor measurements.

Greater source type specificity can be obtained by appropriate selection of sampling locations and periods. Brook et al.⁶⁰ contrasted late winter with summer measurements at an urban and nonurban site. The urban vegetative burning/cooking contribution was much higher during the spring than during the summer, and the absolute spring contribution was much higher at the urban than at the nonurban site. This implies that most of these contributions were from residential wood burning and cooking. During the summer, the absolute vegetative burning contribution in the city was much lower than during the spring, and about half that seen in the nonurban area where agricultural burning is practiced. The summertime urban/nonurban contrast for secondary nitrate and sulfate in eastern Canada shows that most of the nitrate originates within the city, whereas most of the sulfate is distributed throughout the region.

Watson et al.⁹⁹ and Magliano et al.⁴⁸ sampled during morning rush hours and at night, finding higher vehicle exhaust contributions during morning and higher vegetative burning contributions at night. Both of these studies sampled only when PM_{2.5} concentrations were projected to be high on the basis of meteorological forecasts. They took more samples than were intended for analysis and selected a subset after PM_{2.5} and PM₁₀ mass measurements were examined to identify episodes. Both of these measurement approaches conserved substantial resources while focusing the CMB analyses on the high concentration periods of greatest interest. Schauer and Cass⁴⁹ and Pinto et al.⁶² also contrasted

(89) Lundgren, D. A. Regarding sulfur rulemaking: Summary report on the dust emission factor for west prilled sulfur. Prepared for de la Parte and Gilbert, P. A., Tampa, FL, by Environmental Engineering Consultants: Gainesville, FL, 1984.

(90) Midwest Research Institute. Measurement of fugitive dust emissions from prilled sulphur handling; Report No. 7995-L. Prepared for Gardiner, Inc., Tampa, FL, by Midwest Research Institute: Kansas City, MO, 1984.

(91) Corbett, J. J.; Fischbeck, P. S.; Pandis, S. N. *J. Geophys. Res.* **1999**, *104* (D3), 3457-3470.

(92) Corbett, J. J.; Fischbeck, P. S. *Environ. Sci. Technol.* **2000**, *34* (15), 3254-3260.

(93) Davis, D. W.; Hsiao, K.; Ingels, R. M.; Shikiya, J. *JAPCA* **1988**, *38* (9), 1152-1157.

(94) Loranger, S.; Zayed, J. *JAWMA*, **1997**, *47* (9), 983-989.

(95) Zayed, J.; Hong, B.; L'espérance, G. *Environ. Sci. Technol.* **1999**, *33* (19), 3341-3346.

(96) Crump, K. S. *J. Expo. Anal. Environ. Epidemiol.* **2000**, *10* (3), 227-239.

(97) Ressler, T.; Wong, J.; Roos, J.; Smith, I. L. *Environ. Sci. Technol.* **2000**, *34* (6), 950-958.

(98) Truex, T. J.; Pierson, W. R.; McKee, D. E.; Shelef, M.; Baker, R. E. *Environ. Sci. Technol.* **1980**, *14*, 1121-1124.

(99) Watson, J. G.; DuBois, D. W.; DeMandel, R.; Kaduwela, A. P.; Magliano, K. L.; McDade, C.; Mueller, P. K.; Ranzieri, A. J.; Roth, P. M.; Tanrikulu, S. Field program plan for the California Regional PM_{2.5}/PM₁₀ Air Quality Study (CRPAQS). Prepared for California Air Resources Board, Sacramento, CA, by Desert Research Institute: Reno, NV, 1998.

episodes with different meteorology and emissions situations. Schauer and Cass⁴⁹ extracted several filters from entire multi-day episodes to conserve resources for expensive organic compound analyses.

Source Profiles. Many of these studies characterized sources specific to the areas and time periods under study, as noted above. These are valuable new contributions, as too many of the studies relied on measurements made elsewhere or on antiquated profiles reported in the U.S. EPA's SPECIATE database.⁷⁹ Although the SPECIATE database is dated 1999, and provides an interface for identifying, examining, and formatting source profiles, its contents have not been significantly updated since the early work of Shareef et al.¹⁰⁰ and Core et al.¹⁰¹ The point source profiles in the current SPECIATE database are dominated by tests in the late 1970s through the mid-1980s from the U.S. Pacific Northwest,⁸⁵ California,^{102–105} and Philadelphia, PA.¹⁰⁶ Many of these may be somewhat transferable to pre-control conditions such as those experienced by Vega et al.,⁶¹ but they are not applicable to more modern emitters. SCAQMD⁵¹ used the residual oil combustion profile from Watson et al.⁸⁰ that was extracted from crude oil combustion in 1988. Residual oil is no longer used as a fuel in the Los Angeles area, and primary source contributions of Ni and V that indicate this source are probably from refineries and off-shore shipping that have more efficient combustion processes and a higher content of these materials in their profiles. Higher species-to-mass ratios in a source profile result in lower mass contributions from sources represented by that profile.

The observation of Chen et al.⁷² that "The use of widely accessible EPA library profiles is inevitable" is, unfortunately, true. SPECIATE profiles were used to represent coal- and oil-fired power station emissions for Vancouver, BC, in Canada; coal-fired electrical genera-

tion in Taiwan; residual oil combustion in Santa Barbara and Los Angeles, CA, Los Angeles, Mexico City, Korea, and Taiwan; smelting in Mexico City and Korea; diesel engine exhaust in Korea; gasoline and diesel vehicle exhaust in Taiwan, and soil dust in Korea. Where SPECIATE profile numbers were given in the reviewed CMB applications they were traced to the original references that are cited in Table S1 of Supporting Information. Several of the CMB applications in Table S1 did not specify the profile number and it is impossible to judge how well the profiles represented situations in the areas and times of CMB application.

Using profiles from other study areas and times in a CMB source apportionment is reasonable for fossil fuel sources when there are similarities between the fuels, combustion processes, and pollution control measures. This profile justification was not found in most of the applications reviewed here. Source profiles from other times and places might also be used to evaluate primary particle contributions from controlled industrial sources; the source contribution estimates may have large standard errors, but they are often less than 1% to 2% of PM_{2.5} or PM₁₀ mass, as shown in Table S1 of Supporting Information. Even at the upper range of a large relative uncertainty interval, a negligible source contribution is still negligible and this information is adequate for decision-making. As the source contribution estimate becomes larger, however, and the decision based on its value is more expensive, a more region- and time-specific profile becomes more valuable.

Chen et al.⁶⁶ obtained much better results with local fugitive dust profiles than with SPECIATE dust profiles, similar to the situation in the United States.^{108–110} Local paved and unpaved road dust, construction dust, and soil dust profiles should be a prerequisite for receptor modeling because samples are inexpensive to obtain, sieve, resuspend, draw through size-selective inlets onto filters, and quantify for the same substances measured in ambient samples.^{111,112} Fugitive dust profiles measured in the U.S. Pacific Northwest that dominate the SPECIATE database are unlikely to represent these sources in Taiwan or elsewhere. Since fugitive dust profiles contain so many trace elements specific to an area, including those deposited by nearby fossil fuel combustion sources, their profiles can substantially bias source contribution estimates when they are not specific to the ambient sampling period.

The OC and EC carbon fractions specified in source profiles are operationally defined and are not necessarily equivalent among source profiles and receptor measure-

(100) Shareef, G. S.; Bravo, L. A.; Stelling, J. H. E.; Kuykendal, W. B.; Mobley, J. D. Air emissions species database. In *Transactions: Receptor Models in Air Resources Management*; Watson, J. G., Ed.; Air & Waste Management Association: Pittsburgh, PA, 1989; pp 73–83.

(101) Core, J. E.; Rau, J. A.; Chow, J. C.; Watson, J. G.; Pritchett, L. C.; Frazier, C. A.; Kalman, D.; Houck, J. E.; Ward, D.; Cooper, J. A.; Redline, D. Receptor modeling source profile development for the Pacific Northwest States: The Pacific Northwest Source Profile Library. Prepared by Oregon Department of Environmental Quality: Portland, OR, 1989.

(102) Ahuja, M. S.; Paskind, J. J.; Houck, J. E.; Chow, J. C. Design of a study for the chemical and size characterization of particulate matter emissions from selected sources in California. In *Transactions, Receptor Models in Air Resources Management*; Watson, J. G., Ed.; Air & Waste Management Association: Pittsburgh, PA, 1989; pp 145–158.

(103) Cooper, J. A.; Redline, D.; Sherman, J. R.; Valdovinos, L. M.; Miller, E. A.; Pollard, W. L. South Coast Air Basin source profile library; Watson, J. G., Ed.; Air Pollution Control Association: Pittsburgh, PA, 1988.

(104) Houck, J. E.; Chow, J. C.; Watson, J. G.; Simons, C. A.; Pritchett, L. C.; Goulet, J. M.; Frazier, C. A. Determination of particle size distribution and chemical composition of particulate matter from selected sources in California; Report No. A6-175-32. Prepared for California Air Resources Board, Sacramento, CA, by OMNI Environmental Services Inc. and Desert Research Institute: Beaverton, OR, and Reno, NV, 1989.

(105) Houck, J. E.; Goulet, J. M.; Chow, J. C.; Watson, J. G.; Pritchett, L. C. Chemical characterization of emission sources contributing to light extinction. In *Transactions, Visibility and Fine Particles*; Mathai, C. V., Ed.; Air and Waste Management Association: Pittsburgh, PA, 1990; pp 437–446.

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(107) Watson, J. G.; Chow, J. C.; Lu, Z.; Fujita, E. M.; Lowenthal, D. H.; Lawson, D. R. *Aerosol Sci. Technol.* **1994**, *21*, 1–36.

(108) Batterman, S. A.; Dzubay, T. G.; Baumgardner, R. E. *Atmos. Environ.* **1988**, *22* (9), 1821–1828.

(109) Vermette, S. J.; Irvine, K. N.; Drake, J. J. *Environ. Monitoring Assessment* **1991**, *18*, 69–77.

(110) Vermette, S. J.; Williams, A. L.; Landsberger, S. PM₁₀ source apportionment using local surface dust profiles: Examples from Chicago. In *Transactions, PM₁₀ Standards and Nontraditional Particulate Source Controls*; Chow, J. C., Ono, D. M., Eds.; Air & Waste Management Association: Pittsburgh, PA, 1992; pp 262–271.

(111) Chow, J. C.; Watson, J. G.; Houck, J. E.; Pritchett, L. C.; Rogers, C. F.; Frazier, C. A.; Egami, R. T.; Ball, B. M. *Atmos. Environ.* **1994**, *28* (21), 3463–3481.

(112) Carvacho, O. F.; Ashbaugh, L. L.; Matsumura, R. T.; Southard, R. J.; Flochini, R. G. Measurement of PM₁₀ potential from agricultural soils using a dust resuspension test chamber. Proceedings, International Conference on Air Pollution from Agricultural Operations; Midwest Plan Service: Kansas City, MO, 1996.

ments. Several carbon comparison studies^{113,114} show that thermal combustion methods in common use for PM_{2.5} and PM₁₀ on quartz fiber filters yield the same results for total carbon, but different results for OC and EC (also termed BC or soot by some analyses). The distinction between OC and EC by different analysis methods depends on the temperatures at which carbon is evolved, the analysis time at each temperature, the atmosphere surrounding the sample, and optical adjustments for charring of organic carbon at high temperatures in a non-oxidizing atmosphere.⁷⁷ Most of the profiles published in the EPA SPECIATE library measured OC and EC using the thermal optical reflectance method developed by Huntzicker et al.¹¹⁵ that is equivalent to the IMPROVE method.¹¹⁶ When OC/EC fractions are measured by methods that differ from those of the profiles, source contribution estimates that rely on OC and EC abundances will be incorrect. Watson et al.¹⁰⁷ found this to be the case for CMB source apportionment in the South Coast Air Basin when OC/EC source profiles from Cooper et al.^{103,117} were found incompatible with ambient OC/EC measurements made by the method of Fung.¹¹⁸ Only total carbon could be used in the CMB, with the result that vegetative burning/cooking was attributed to the vehicle exhaust profile. Chow et al.⁷⁷ show that an additional 850 °C temperature added during the oxygen-free portion of the NIOSH¹¹⁹ thermal/optical analysis protocol produces EC measurements that are often half those of the IMPROVE method. NIOSH¹¹⁹ intends for this high temperature to represent a carbonate interference with diesel exhaust that might be found in underground mines, but Chow and Watson¹²⁰ find that there is little carbonate in ambient air samples. Chow et al.⁷⁷ show that substantial amounts of EC evolve at this high temperature, even in the absence of oxygen.

Carbon fractions that evolve at different temperatures and with different optical properties could be used to advantage for CMB source apportionment if measured consistently at both source and receptor. Watson et al.¹²¹ showed substantial differences in fractions of carbon concentrations as a function of temperature for gasoline and diesel vehicle exhaust, while Engelbrecht et al.⁶³

used differences in four OC and three EC fractions to distinguish D-grade coal versus synthetic low-smoke fuel source contribution estimates. Jeon et al.¹²² take this a step further by directing the effluent from heated filters through a gas chromatograph with mass spectrometric detection. It is not necessary that these carbon fractions are identifiable as specific chemical species for CMB source apportionment. They must only be consistently measured in source and receptor samples and meet the other requirements to eliminate collinearity between source profiles.

Some organic compound source profiles have been measured for gasoline and diesel vehicle exhaust,^{123–128} meat cooking,^{128–131} natural gas combustion,¹³² coal combustion,¹³³ oil combustion,¹³⁴ wood burning and forest fires,^{135–140} and fugitive dust.¹⁴¹ Most of these results are from emitters in southern California with some from Denver, CO. Different investigators measure and use different markers for these sources in receptor models. The same measurements, which are often very costly, are also needed at the receptors. This technology is rapidly evolving and may soon be in widespread use.

Simoneit¹⁴² cited several examples of odd- and even-numbered carbon molecules in the *n*-alkane series as indicating the presence or absence of organic carbon from man-made sources in the presence of ubiquitous contributions from natural sources. Biogenic material, such as plant waxes, tend to have more molecules with odd-numbers of carbon atoms, whereas *n*-alkanes from combustion processes have nearly equal quantities of

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even and odd carbon-numbered molecules. A Carbon Preference Index (CPI), the ratio of odd to even *n*-alkane masses, is used to estimate the relative abundances of odd and even compounds to assess the relative abundance of natural vs anthropogenic contributions.

Performance and Evaluation. Watson et al.^{16,17} specify an applications and validation protocol that was rarely followed in the reviewed CMB applications. The seven steps in this protocol are the following: (1) determine model applicability on the basis of availability of input data; (2) select and justify profiles to represent identified contributors; (3) evaluate model outputs and performance measures; (4) identify and evaluate deviations from model assumptions; (5) identify and correct input data deficiencies; (6) verify consistency and stability of source contribution estimates; and (7) evaluate CMB results with respect to other data analysis and source assessment methods.

Several of the studies summarized performance statistics, but most did not. Schauer et al.,⁴⁵ Fujita et al.,¹⁴³ and Watson et al.¹⁰⁷ provide the best examples of sensitivity testing and source evaluation.

Documentation. Lack of documentation is a major shortcoming for all of these studies. This is especially the case for source profiles that should specify where they were obtained, how they relate to the sources under study, equivalence to receptor measurements (especially OC/EC), and emissions within the urban or regional domain that might contribute. Part of this is due to limitations on publication lengths. This could be alleviated by making better use of the web-based "Supplemental Information" supported by American Chemical Society publications. At the very least, CMB input files could be archived as supplemental data.

Summary and Conclusions

The CMB receptor model serves several objectives in air quality assessment. It directly apportions ambient

PM_{2.5} and PM₁₀ concentrations to their primary sources. This apportionment in and of itself has been of great utility in developing control strategies in PM₁₀ nonattainment areas over the past decade. The CMB has been the primary analysis tool in many locations that have no emissions inventories. The CMB independently verifies emissions inventories, against which emissions reduction credits are taken, so that expenditures on their improvement may be better focused. The CMB also has the potential to estimate contributions of different source types to secondary sulfates and nitrate, especially when coupled with source-oriented models that can simulate changes in source profiles during transport between source and receptor.

CMB analyses in 22 different studies have found fossil fuel combustion to be a large contributor to PM_{2.5} and PM₁₀ concentrations, with most of the primary contributions originating from gasoline- and diesel-powered vehicle exhaust. Primary contributions from ducted sources, such as coal- and oil-fired power stations, are negligible when these facilities have been modernized with effective pollution controls, but they have been shown to be large contributors without these controls. Secondary sulfates and nitrates from fossil fuel combustion are recognized, but their contributions are uncertain from both the CMB and source models. Using source and receptor models together improves source contribution estimates and the confidence in those estimates that can be used by decision-makers to implement costly emissions reduction strategies.

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Supporting Information Available: Summary of PM source apportionment studies using CMB and other receptor models. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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