

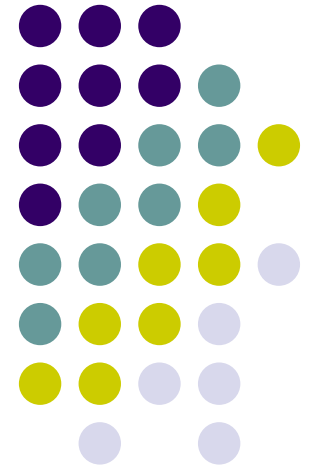


Chemical Mass Balance Application

Alan Gertler

November 2005

CMB Application

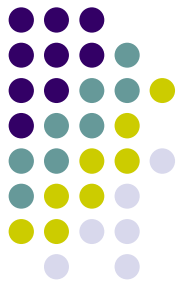


Topics



- CMB Model
- Examples of sources of input data (Note – you will generate your own data for Hyderabad)
- Protocol for applying and validating the model
- Additional resources

Chemical Mass Balance



Equation:
$$C_j = \sum_{j=1}^J F_{ij} S_j \quad \text{for } i = 1 \text{ to } N$$

Input:

- Ambient concentrations (C_i) and uncertainties (σ_{Cj}), source profiles (F_{ij}), and uncertainties (σ_{Fij}).

Output:

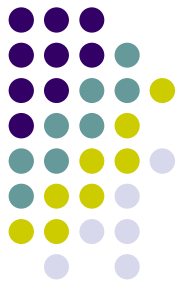
- Source contributions (S_j) and uncertainties (σ_{Sj}).

Measurements:

- Size-classified mass, elements, ions, and carbon concentrations on both ambient and source samples.

CMB Solutions

Minimize differences between calculated and measured values for overdetermined set of equations



$$\chi^2 = \min \sum_i [(C_i - \mathbf{C}_i)^2 / \sigma_{C_i}^2] + \sum_i \sum_j [(F_{ij} - \mathbf{F}_{ij})^2 / \sigma_{F_{ij}}^2]$$

Britt and Luecke, (1973), single sample, bold=true value

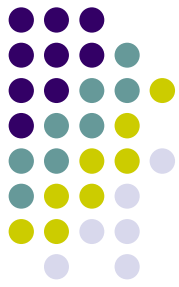
$$\chi^2 = \min \sum_i [(C_i - \sum_j F_{ij} S_j)^2 / (\sigma_{C_i}^2 + \sum_j \sigma_{F_{ij}}^2 S_j^2)]$$

Effective Variance, Watson et al., (1984), single sample

$$\chi^2 = \min \sum_i [(C_i - \sum_j F_{ij} S_j)^2 / \sigma_{C_i}^2]$$

Ordinary Weighted Least Squares, Friedlander (1973), single sample

Other CMB Solutions



$$S_j = C_i / F_{ij}$$

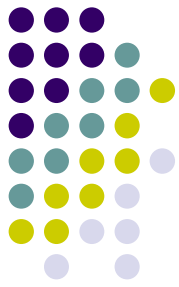
Tracer solution, Hidy and Friedlander (1971), Winchester and Nifong (1971), single sample

$$\chi^2 = \min \sum_k [(Mass_k - \sum_i C_{ik} / F_{ii})^2]$$

Multiple Linear Regression, Kleinman et al (1980), multiple samples

$$\chi^2 = \min \sum_i \sum_k [(C_{ik} - \sum_j F_{ij} S_{jk})^2 / \sigma_{C_{ik}}^2]$$

Positive Matrix Factorization, Paatero (1997), multiple samples



Time-Integrated Sampling

Airmetrics portable
MiniVol sampler

BGI FRM Omni

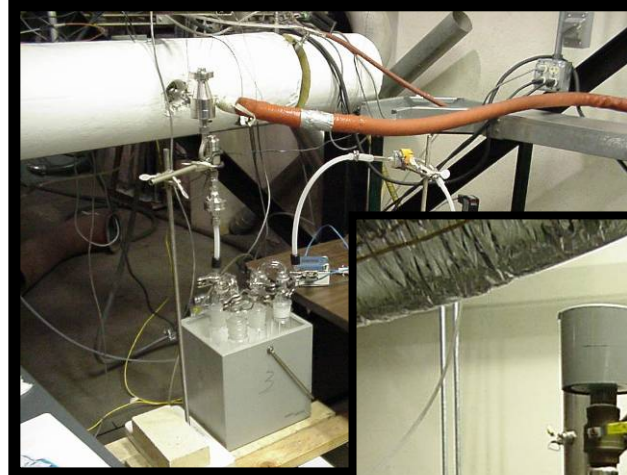


PM_{2.5} and PM₁₀

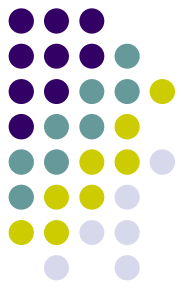


PM₁, PM_{2.5}, and PM₁₀

Dilution Testing of Foundry Emissions



Source-Dominated Sampling (Cooking)



Real-World Cooking



Simulated Cooking

Inspection and Maintenance Compliance Tests

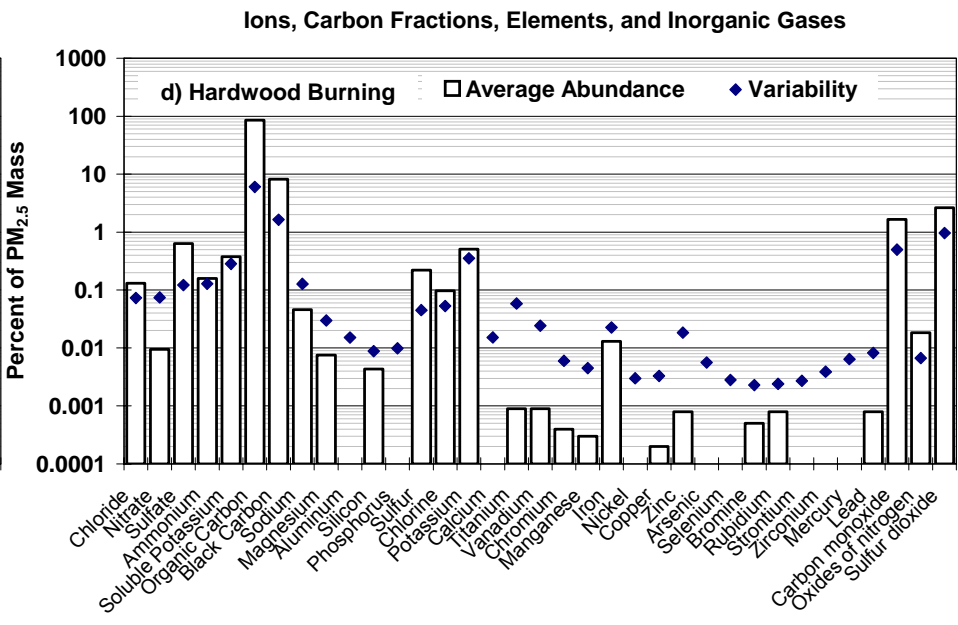
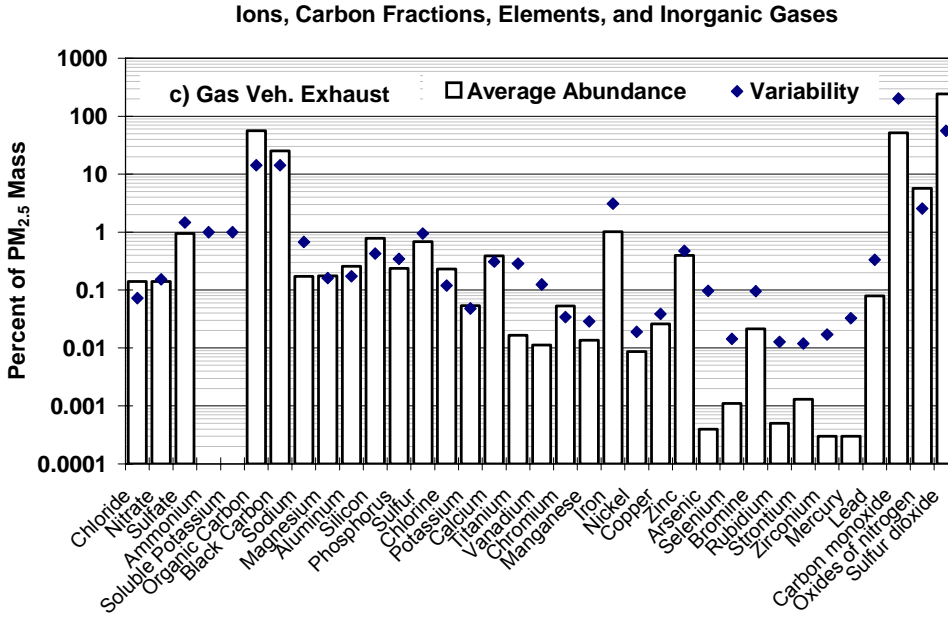
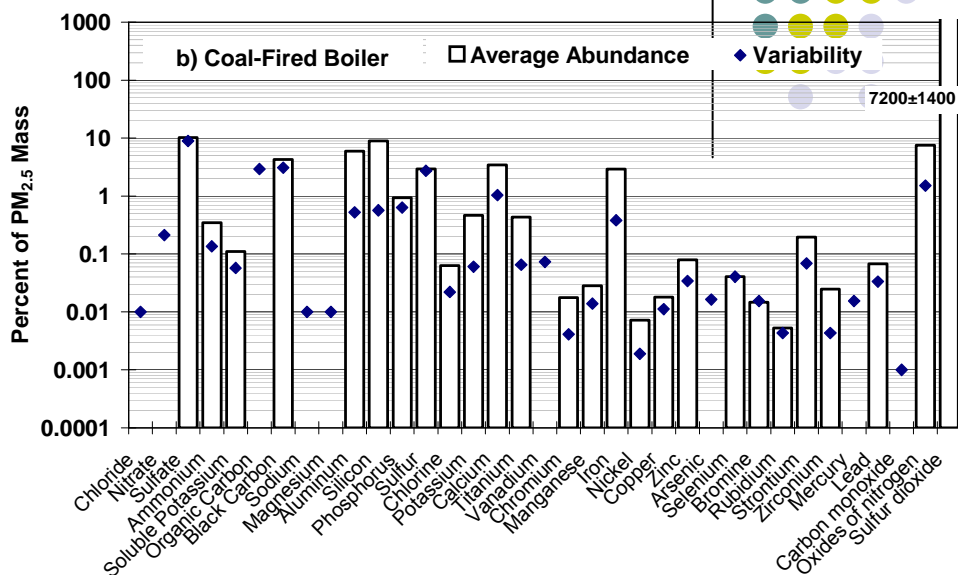
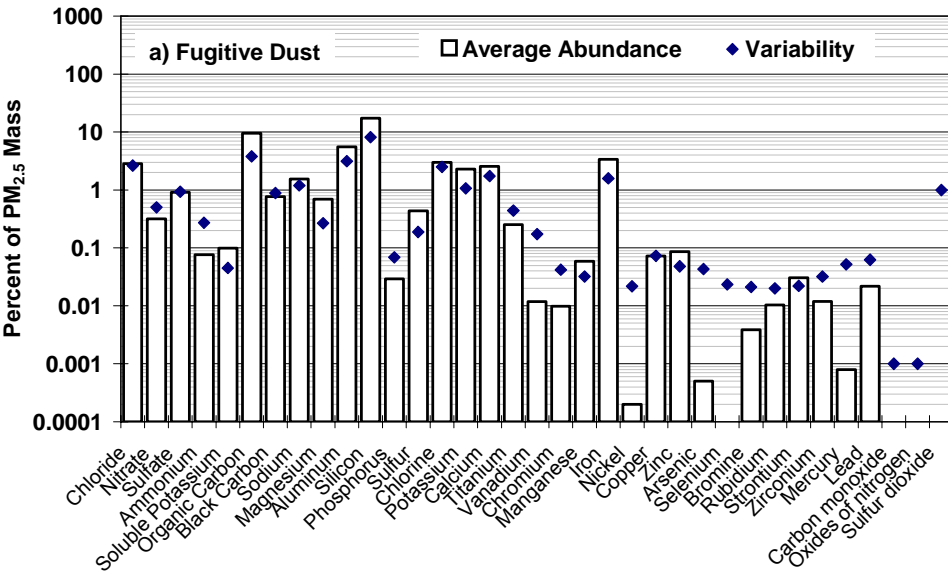


Roadside compliance test in India



Source Profiles

Commonly measured elements, ions, carbon (Zielinska et al., 1998)



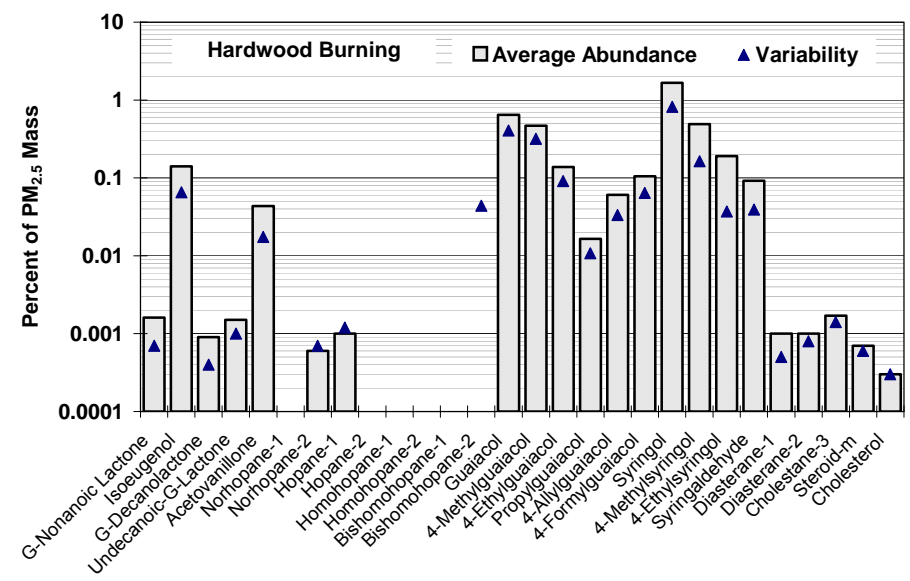
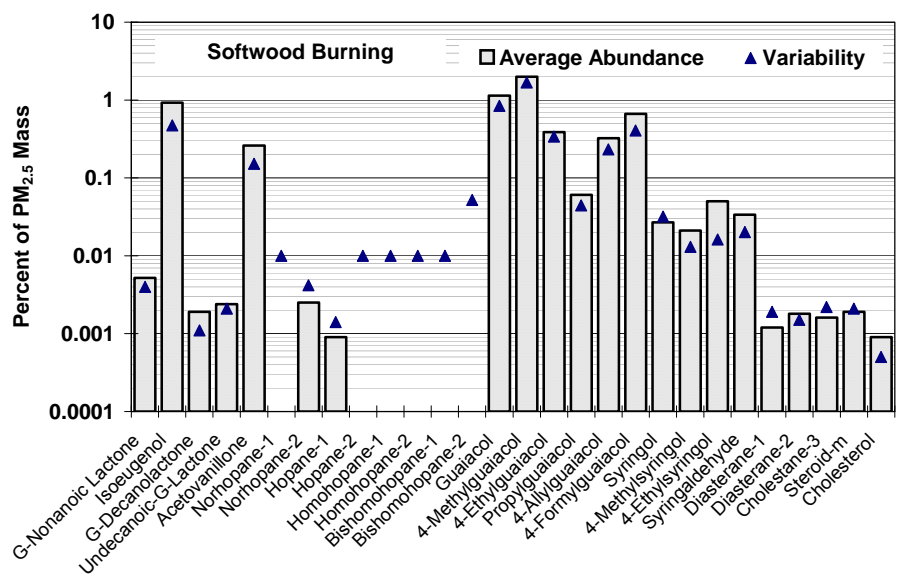
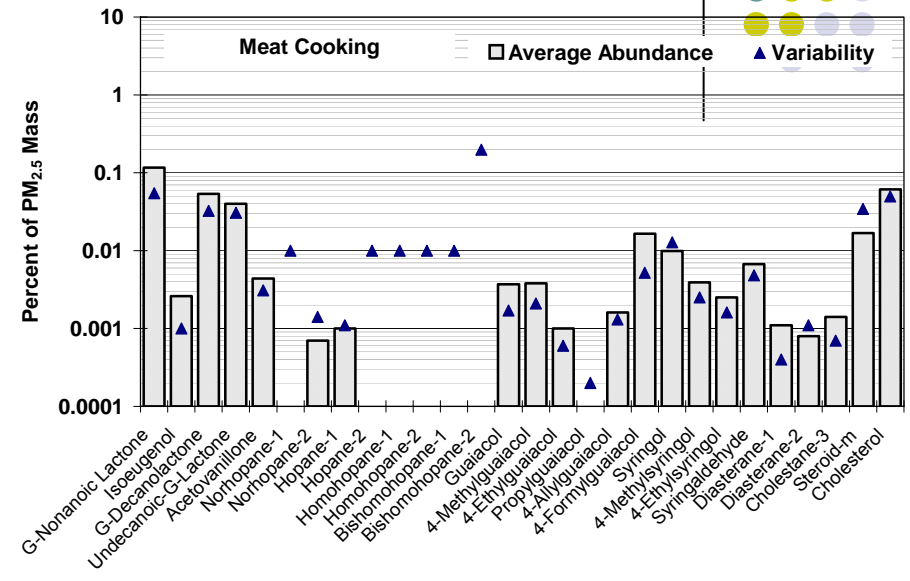
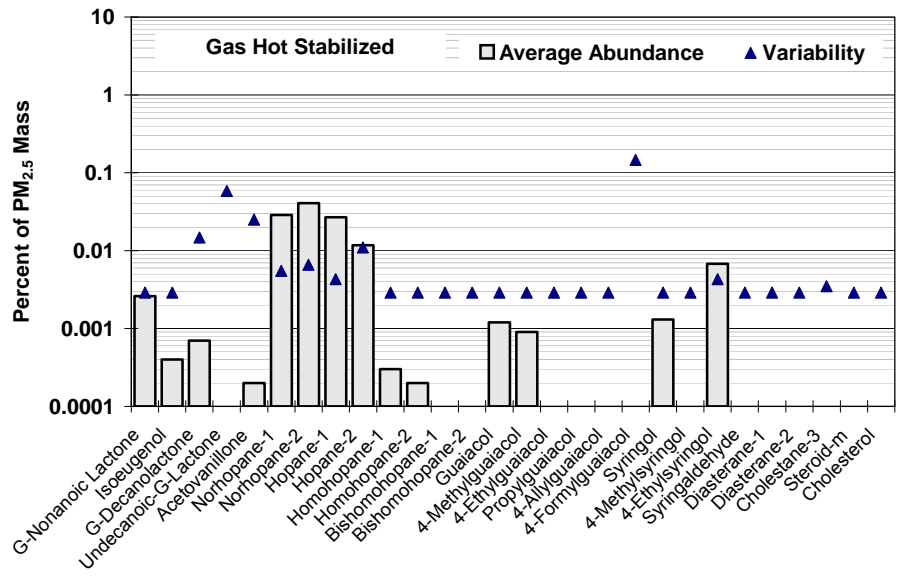
Ions, Carbon Fractions, Elements, and Inorganic Gases

Ions, Carbon Fractions, Elements, and Inorganic Gases

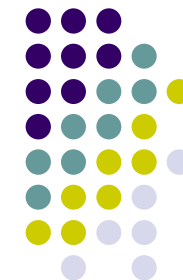
Organic Source Profiles Better Distinguish Among Sources

(lactones, hopanes, guaiacols, syringols, steranes, and sterols)

Zielinska et al. (1998)



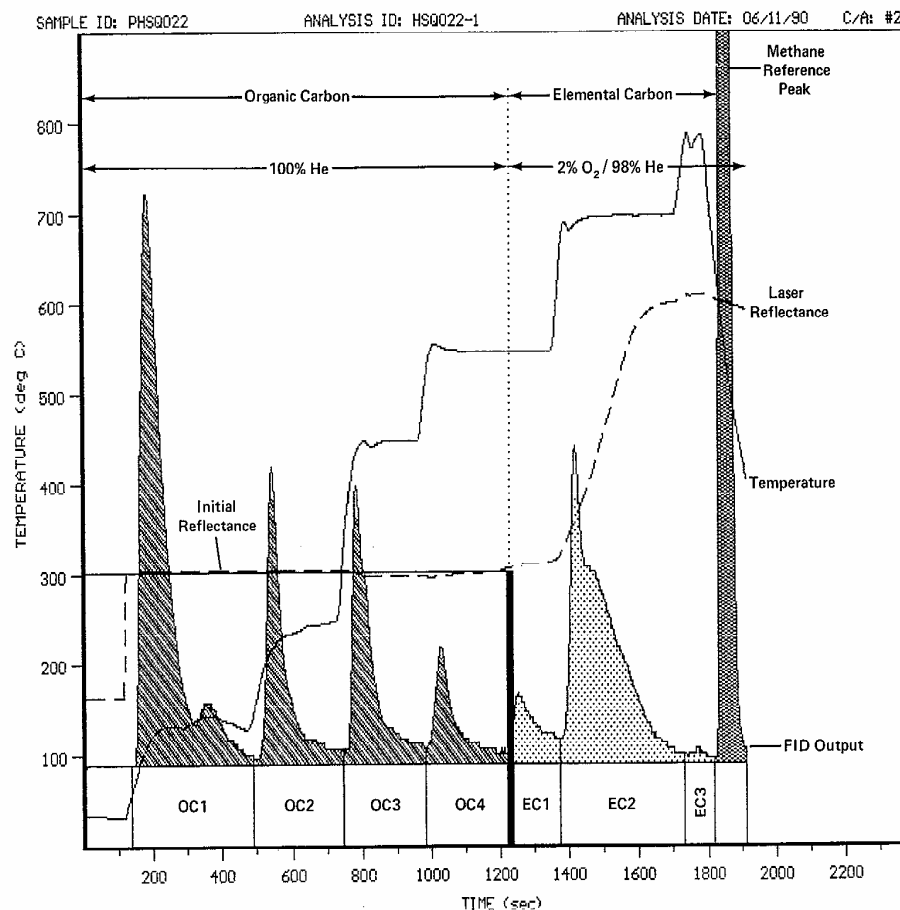
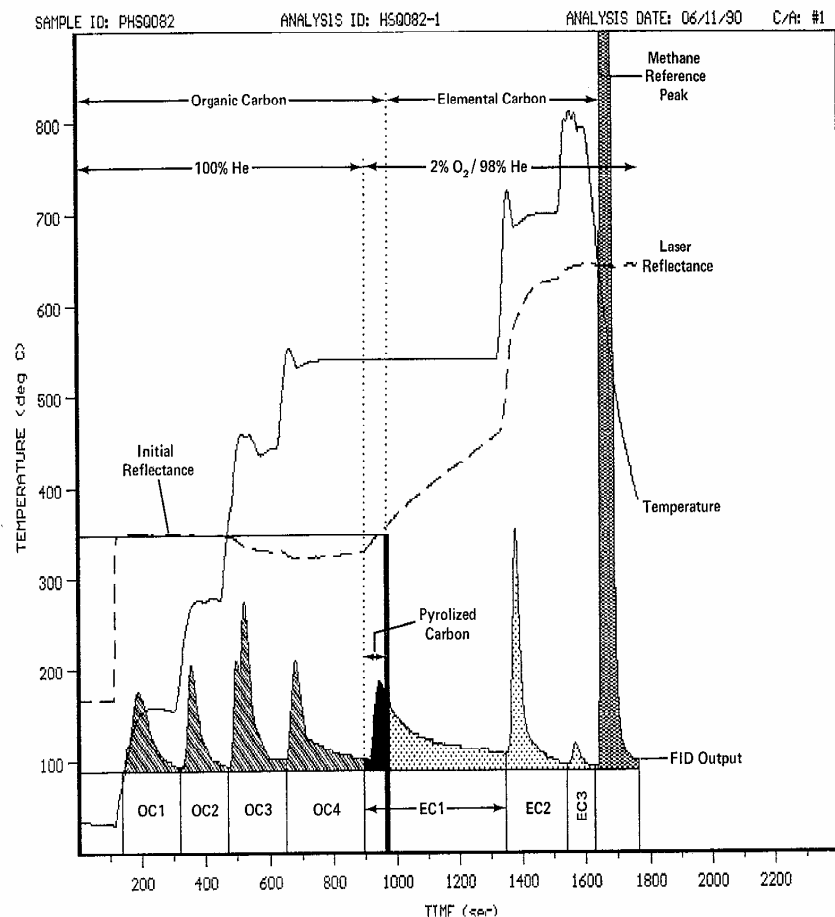
Source Properties can be Operationally Defined



(Thermally evolved carbon fractions, Watson et al., 1994)

Gasoline-fueled vehicles

Diesel-fueled vehicles

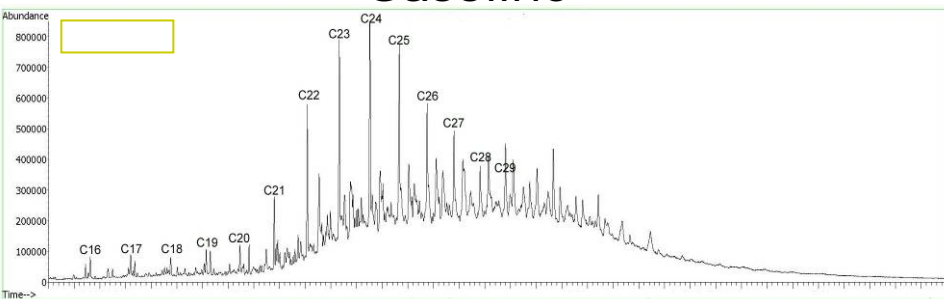


Organic Compounds and Patterns can be Measured on Small Samples

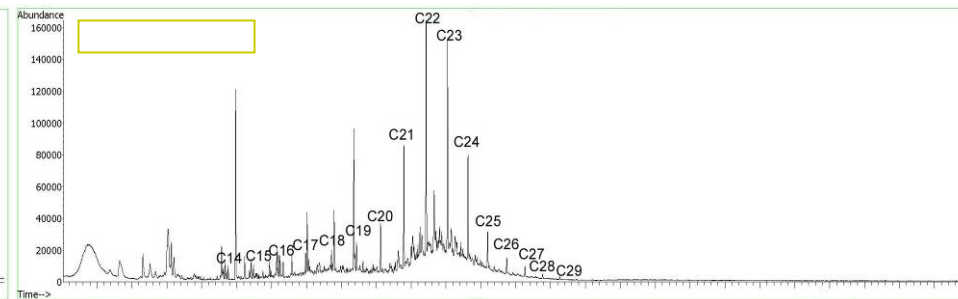
(Example* Chromatograms of Thermal Desorption GC/MS)



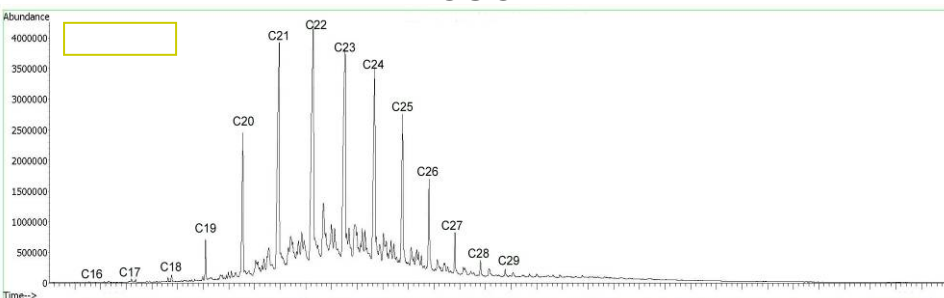
Gasoline



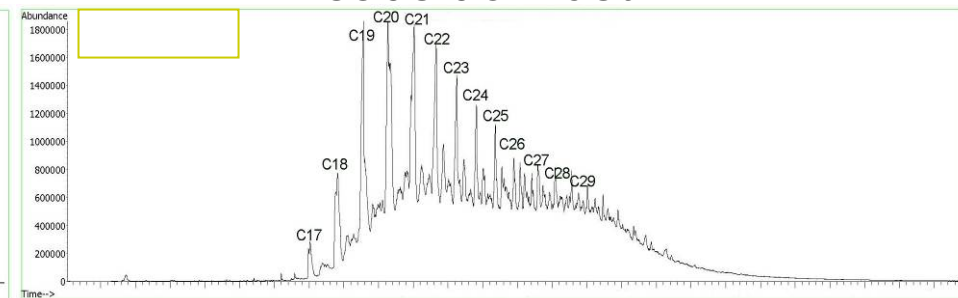
Coal Power Plant



Diesel

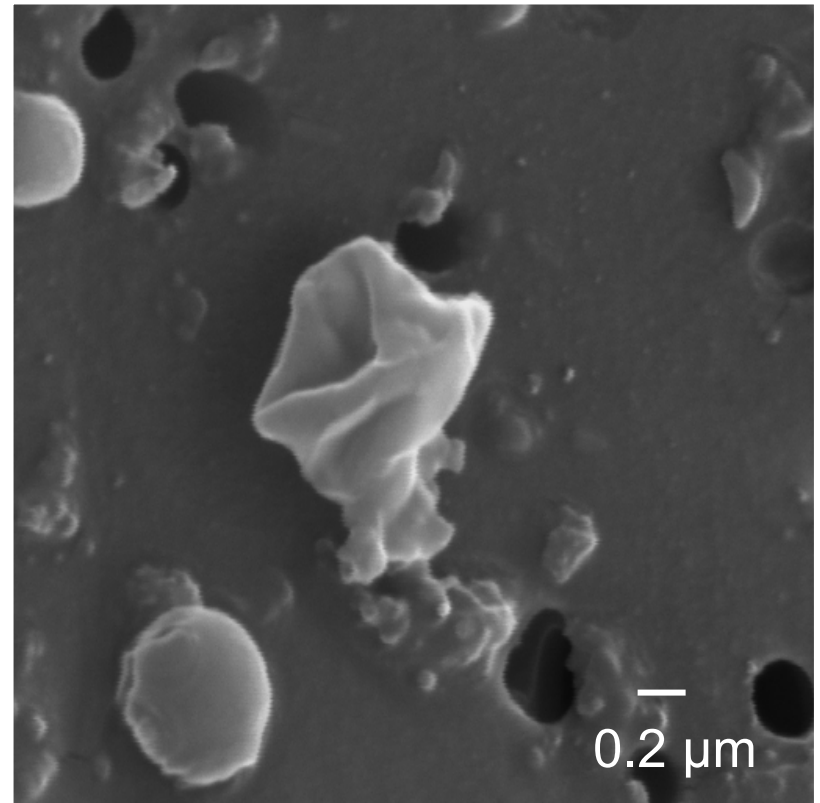
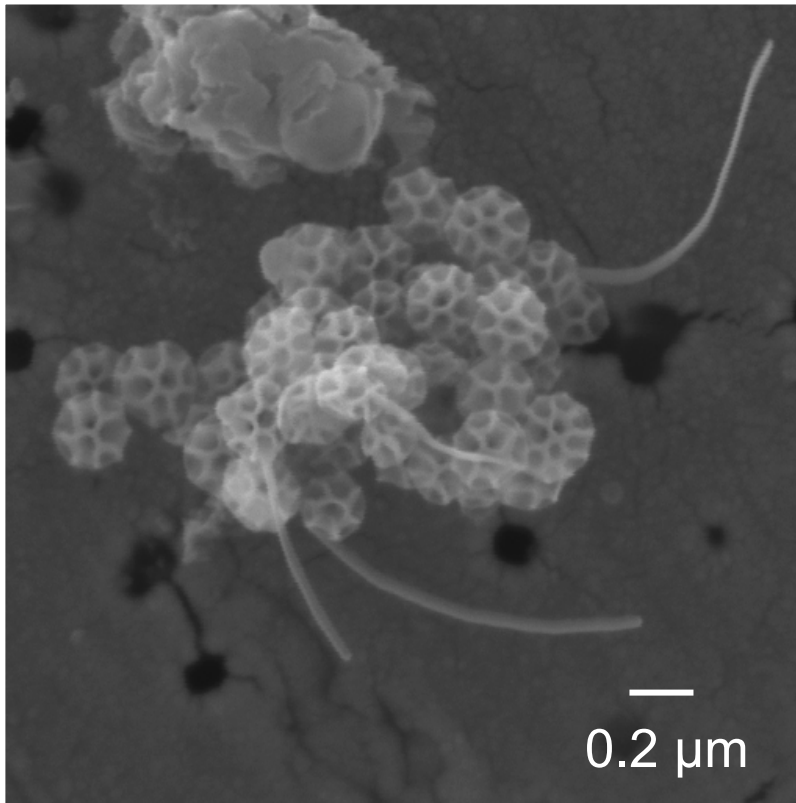
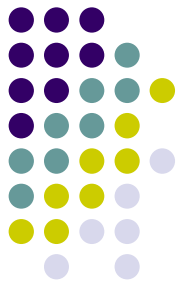


Roadside Dust



* Ion 57 Chromatogram for alkanes

Microscopic Analysis

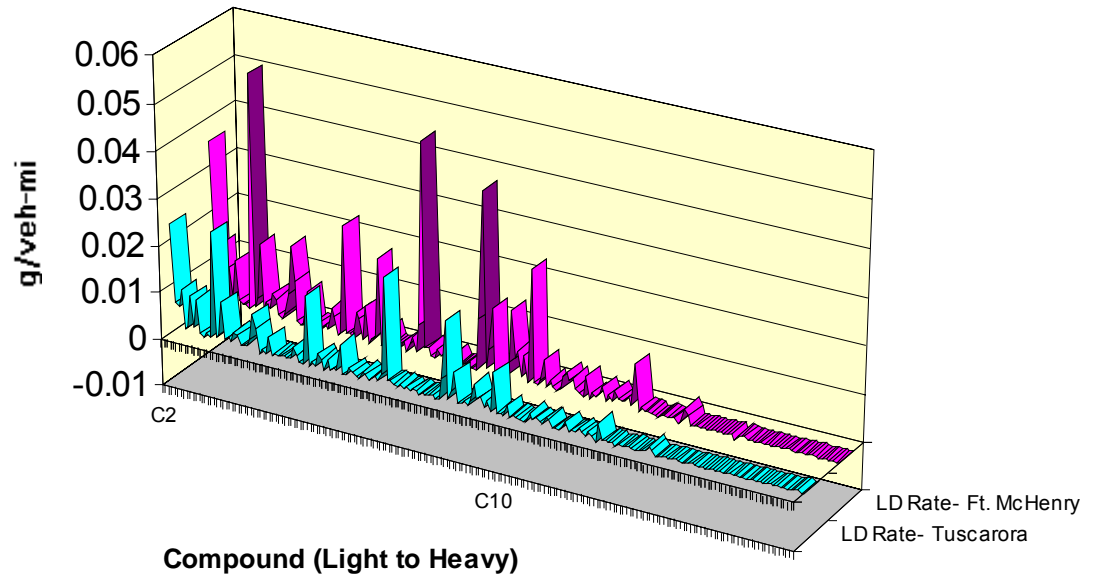




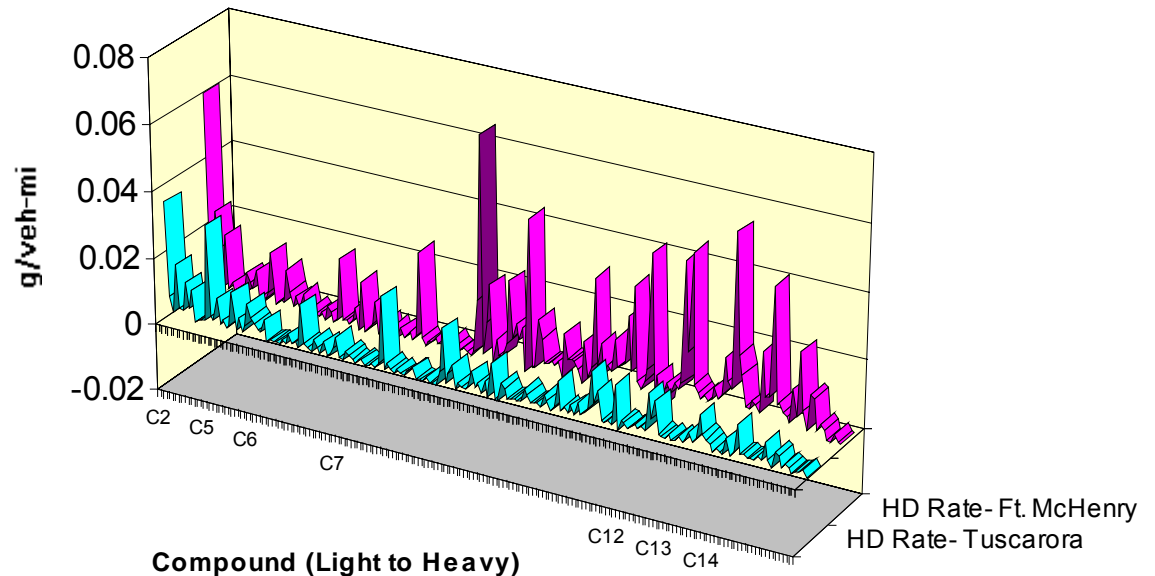
Gases can be Included in Profiles

(Gertler et al., 1996)

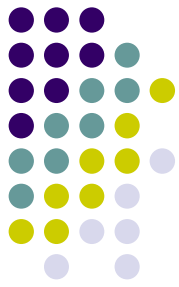
Light Duty Emission Rates



Heavy Duty Emission Rates



Source Composition Data Needs



- Profiles for desired source types
- Marker properties (elements, ions, carbon fractions, organic compounds, isotopic abundances, single particle properties)
- Same particle size ranges and species as measured at receptor
- Measurement methods equivalent to those of receptor samples
- Documentation of source characteristics, fuels, and operating parameters
- Source profile uncertainties
- Profiles are as they would appear at the receptor

Source Profile Availability



- Published articles and reports
- U.S. EPA SPECIATE data base
- Researcher data bases
- Original measurements
 - Hot stack compliance sampling (ducted sources, misses condensed species)
 - Dilution stack sampling (ducted sources, more realistic of what is at receptor)
 - Laboratory simulation (dynamometers, test combustors, smog chambers)
 - Source dominated sampling (tunnel, roadside, wildfire plume)
 - Resuspension sampling (fugitive dust, hopper ash, road salt)

Ambient Data Availability

<http://vista.cira.colostate.edu/views/Web/QueryWizard/QueryWizardClient.aspx>

<http://www.arb.ca.gov/airways/Datamaintenance/default.asp>

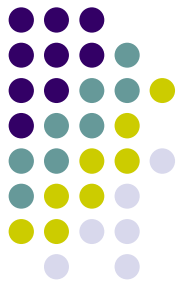


The screenshot shows the 'Visibility Database Query Wizard (VIEWS)' web application running in Microsoft Internet Explorer. The browser's address bar displays the URL: <http://vista.cira.colostate.edu/views/Web/QueryWizard/QueryWizardClient.aspx>. The application interface is divided into several sections:

- SELECT MONITORING LOCATIONS:** This section contains two lists for selection. The 'Select Networks:' list includes ARS, CASTNet, EPAFRM, EPASPEC, IMPROVE, MOHAVE, NESCAUM, PREVENT, REVEAL, SEAVS, and SFU. The 'Select Sites:' list includes various locations such as ARS AZ: Humble Mountain (Pine Mnt Wilderness Area), ARS AZ: Mazatzal, ARS AZ: McFadden Peak, ARS AZ: Muleshoe Ranch, ARS AZ: Phoenix, ARS AZ: Phoenix (Transmissometer), ARS AZ: Rucard Canyon (Chiricahua), ARS AZ: Sierra Ancha, ARS AZ: Sycamore Canyon Wilderness Area, ARS AZ: Tucson, ARS AZ: Tucson Central, ARS AZ: Tucson Mountain, ARS CO: Fort Collins, ARS WA: Mount Saint Helens, and ARS WA: North Cascades. There is also a 'Select by RPD:' section with checkboxes for WRAP, CENRAP, Midwest-RPD, MANE-VU, and VISTAS.
- SELECT DATES FOR WHICH TO RETRIEVE DATA:** This section offers two options: 'By Years and Months:' and 'By Date Ranges:'. Under 'By Years and Months:', there are two lists: one for years (2002 to 1991) and one for months (January to December). Under 'By Date Ranges:', there are fields for 'Start Date' (January 2002) and 'End Date' (January 2002), along with a 'Add This Range' button and a 'clear' button.
- SPECIFY THE PARAMETERS YOU WISH TO VIEW:** This section is partially visible at the bottom of the screen.

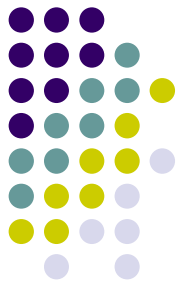
The browser's taskbar at the bottom shows the Start button, several open Microsoft Office applications, and the system clock displaying 11:58 AM on a desktop.

Examples of USA Receptor Model Air Quality Findings and Results



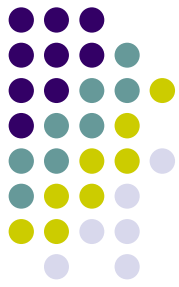
- Oregon wood stove emissions standard (Watson, 1979)
- Midwest contributions to east coast sulfate and ozone (Wolff et al., 1977, Lioy et al., 1980, Mueller et al., 1983, Rahn and Lowenthal, 1984)
- Washoe County, Nevada, stove changeout, burning ban, and “squealer” number (Chow et al., 1989)
- California EMFAC emissions model revisions (Fujita et al., 1992, 1994)
- SCAQMD (Los Angeles) grilling emission standard (Rogge, 1993)
- SCAQMD (Los Angeles) street sweeper specification (Chow et al., 1990)
- SCAQMD (Los Angeles) Chino dairy reduction (NH₃) regulation (SCAQMD, 1996)
- PM₁₀ SIP implementation of wood burning, road dust, and industrial emission reductions (Davis and Maughan, 1984, Houck et al., 1981, 1982, Cooper et al., 1989)
- Navajo Generating Station SO₂ scrubbers (Malm et al., 1989)
- Hayden Generating Station SO₂ scrubbers (Watson et al., 1996)
- Mohave Generating Station shutdown (Pitchford et al., 1999)
- Denver Colorado urban visibility standard (Watson et al., 1988)

The CMB Receptor Model and its derivatives are not Statistical



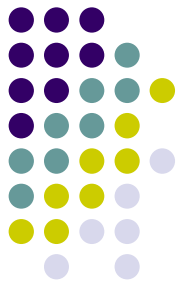
- They don't test hypotheses or determine statistical significance
- They are physically based with statements of simplifying assumptions and evaluation of deviations from assumptions
- They infer mechanisms and interactions rather than explicitly calculate them
- Receptor models recognize and elucidate patterns in measured components, space and time that bound the types, quantities, and locations of source contributions
- Some of them explicitly use input data uncertainties to weight influence of inputs and estimate uncertainties of outputs

Receptor models can get at secondaries under certain conditions



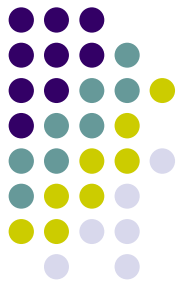
- OC/EC enrichment factors used to estimate secondary OC contributions (Turpin and Huntzicker, 1991, Gray et al., 1986)
- Secondary organic marker end-products (Pandis, 2001)
- Aerosol evolution to represent changes in profiles (Lewis and Stevens, 1985, Watson et al., 2002)
- ^{34}S or ^{35}S isotopes to follow sulfate changes (Forrest and Newmann, 1973, Hidy, 1987)
- Regional source profiles (Rahn and Lowenthal, 1984, Eatough et al., 1997)
- Eigenvector-derived profiles (Poirot et al., 2001)

CMB Model Assumptions



- Compositions of source emissions are constant over the period of ambient and source sampling.
- Chemical species do not react with each other (i.e., they add linearly).
- All sources with a potential for significant contribution to the receptor have been identified and have had their emissions characterized.

CMB Model Assumptions (continued)



- The number of sources is less than or equal to the number of chemical species.
- The source compositions are linearly independent of each other.
- Measurement errors are random, uncorrelated, and normally distributed.



CMB8 APPLICATIONS AND VALIDATION PROTOCOL FOR PM_{2.5} AND VOCS

Desert Research Institute Document No. 1808.2F3

June 29, 2001

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Protocol for Applying and Validating the CMB Model

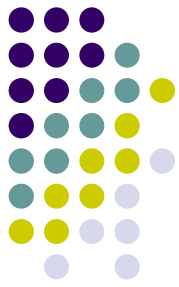


1. Assess general applicability.
2. Configure with source types, source profiles, and chemical species from receptor.
3. Examine model statistics and diagnostics.
4. Determine compliance with model assumptions.

Protocol for Applying and Validating the CMB Model (continued)



5. Modify model configuration to better comply with assumptions.
6. Test the consistency and stability of CMB results.
7. Evaluate the validity of model results.



1. General Applicability

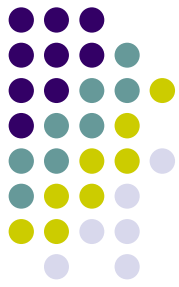
- Samples amenable to or have been chemically speciated
- Potential source contributors identified
- Source profiles measured or approximated
- Marker species in source profiles and receptor measurements
- More receptor species than source types



2. Model Configuration

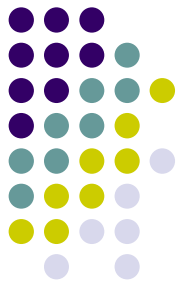
- Receptor species:
 - A value and uncertainty is needed for each species
 - Only one measurement of a given species should be included in the solution (e.g., S and SO₄)
 - Values below lower quantifiable limits may be included if uncertainty is set to LQL

2. Model Configuration (continued)



- Source type selection:
 - Common area sources (vehicle exhaust, geological material, secondary sulfate and nitrate)
 - Natural sources (sea salt)
 - Point sources in emissions inventory (coal combustion, residual oil combustion, smelting)
 - Sources identified in PCA or preliminary analysis

2. Model Configuration (continued)



- Source profiles in CMB solution:
 - Upwind point sources
 - Seasonal emitters
 - Non-collinear profiles



3. Model Outputs, Statistics, and Diagnostics

SOURCE CONTRIBUTION ESTIMATES - SITE: PACS1 DATE: 08/13/77 VERSION: 7.0
 SAMPLE DURATION 24 START HOUR 0 SIZE: FINE
 R SQUARE .98 PERCENT MASS 98.7
 CHI SQUARE 1.12 DF 13

SOURCE	* TYPE	SCE(UG/M3)	STD ERR	TSTAT	
1	MARIN	12.3889	2.2457	5.5167	Marine Aerosol
3	UDUST	9.5917	1.3876	6.9127	Urban Dust
4	AUTPB	10.0835	1.4942	6.7486	Leaded Motor Vehicle Exhaust
5	RDOIL	11.0603	1.9239	5.7490	Residual Oil Combustion
8	KRAFT	4.6896	5.0467	.9292	Kraft Recovery Boiler
11	ALPRO	10.6023	3.5896	2.9536	Aluminum Pot Line
12	STEEL	8.6729	1.3771	6.2979	Steel Blast Furnace
13	FERMN	11.8754	1.8321	6.4820	Ferromanganese Furnace

MEASURED CONCENTRATION FINE/COARSE/TOTAL:
 80.0+- 8.0/ 80.0+- 8.0/ 160.0+- 11.3

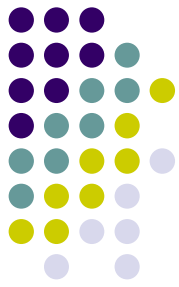
UNCERTAINTY/SIMILARITY CLUSTERS VERSION: 7.0 SUM OF CLUSTER SOURCES

1	8			17.078+-	4.241
1	5	8		28.139+-	3.833

SPECIES CONCENTRATIONS - SITE: PACS1 DATE: 08/13/77 VERSION: 7.0
 SAMPLE DURATION 24 START HOUR 0 SIZE: FINE
 R SQUARE .98 PERCENT MASS 98.7
 CHI SQUARE 1.12 DF 13

SPECIES	I ^a	MEAS ^b	CALC ^b	RATIO C/M	RATIO R/U				
C1	TOT	T	80.00000+-	8.00000	78.96461+-	4.82449	.99+-	.12	-.1
C9	F		.88300+-	.08800	.67644+-	.24792	.77+-	.29	-.8
C11	NA	*	6.93000+-	.69300	6.97025+-	.56446	1.01+-	.13	.0
C12	MG	*	.43000+-	.04300	1.60951+-	.62627	3.74+-	1.50	1.9
C13	AL	*	4.66000+-	.46600	4.02418+-	.88919	.86+-	.21	-.6
C14	SI	*	3.02000+-	.30200	2.92212+-	.13329	.97+-	.11	-.3
C16	S		2.95000+-	.29500	3.02466+-	.31807	1.03+-	.15	.2
C17	CL	*	5.95000+-	.59500	5.69381+-	1.24836	.96+-	.23	-.2
C19	K	*	1.64000+-	.16400	1.73084+-	.46411	1.06+-	.30	.2
C20	CA	*	1.78000+-	.17800	1.43537+-	.11366	.81+-	.10	-1.6
C22	TI	*	.08300+-	.00800	.10088+-	.01630	1.22+-	.23	1.0
C23	V	*	.37200+-	.03700	.39757+-	.08308	1.07+-	.25	.3
C24	CR	*	.31500+-	.03200	.20976+-	.12151	.67+-	.39	-.8
C25	MN	*	2.99000+-	.29900	2.82844+-	.14115	.95+-	.11	-.5
C26	FE	*	4.53000+-	.45300	4.24446+-	.33269	.94+-	.12	-.5
C28	NI	*	.76500+-	.07700	.68246+-	.13428	.89+-	.20	-.5
C29	CU	*	.04400+-	.00400	.05274+-	.00510	1.20+-	.16	1.3
C30	ZN	*	.22500+-	.02300	.26786+-	.03966	1.19+-	.21	.9
C35	BR	*	.41900+-	.04200	.56133+-	.17386	1.34+-	.44	.8
C82	PB	*	2.53000+-	.25300	2.13749+-	.30300	.84+-	.15	-1.0
C201	OC	*	7.54000+-	.75400	8.50978+-	1.35632	1.13+-	.21	.6
C202	EC	*	1.42000+-	.14200	1.33579+-	.34012	.94+-	.26	-.2
C203	SO4	*	10.30000+-	1.03400	9.78819+-	1.47514	.95+-	.17	-.3
C204	NO3	*	.63800+-	.06400	.88402+-	.35938	1.39+-	.58	.7

3. Model Outputs, Statistics, and Diagnostics (cont.)



Source Contribution Display

<u>Output/Statistic/Code</u>	<u>Abbreviation</u>	<u>Description</u>
Source Contribution Estimate	SCE	Primary output: Source contribution ($\mu\text{g}/\text{m}^3$).
Standard Error	STDERR	Primary output: The variance of the SCE. [Target: \ll SCE]
t-statistic	T-STAT	Statistic: The ratio of the SCE to its STDERR. A high T-STAT suggests a non-zero SCE. [Target: > 2.0]
R-square	R-SQUARE	Statistic: Used to measure the variance in the ambient species concentrations which is explained by the calculated species concentrations. Ranges from 0 to 1.0. [Target: 0.8 to 1.0]

3. Model Outputs, Statistics, and Diagnostics (cont.)



Source Contribution Display (continued)

<u>Output/Statistic/Code</u>	<u>Abbreviation</u>	<u>Description</u>
Chi-square	CHI-SQUARE	Statistic: Used to consider the uncertainties of the calculated species concentrations. A high CHI-SQUARE suggests that the model has not explained the species data very well. [Target: 1.0 to 4.0]
Percent Mass Accounted For	PERCENT MASS or % MASS	Statistic: Used to track the % of ambient mass explained by the sum of the SCEs. A % MASS near 100% can be misleading because a poor fit can force a high % MASS. [Target: 100% \pm 20%]

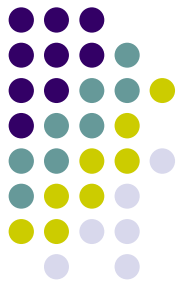
3. Model Outputs, Statistics, and Diagnostics (continued)



Source Contribution Display (continued)

<u>Output/Statistic/Code</u>	<u>Abbreviation</u>	<u>Description</u>
Degrees of Freedom	DF	Statistic: Number of species in fit minus number of sources in fit.
Site/Sample	SITE, etc.	Status: Data being used in the fit.
Uncertainty/Similarity Clusters	U/S Clusters	Diagnostic: Summary of singular value decomposition analysis. It shows clusters of sources which the model cannot easily distinguish between and that are likely to be interfering with the model's ability to provide a good set of SCEs. [Target: No Clusters]

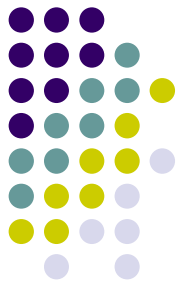
3. Model Outputs, Statistics, and Diagnostics (continued)



Source Contribution Display (continued)

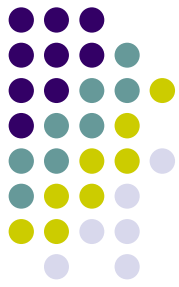
<u>Output/Statistic/Code</u>	<u>Abbreviation</u>	<u>Description</u>
Sum of Combined Sources	SUM \pm	Supplemental output: Supplement to U/S Clusters. It estimates the sum of SCEs of the sources in a cluster and the standard error of the sum. The standard error of the sum follows the \pm .

4. Evaluate Model Assumptions



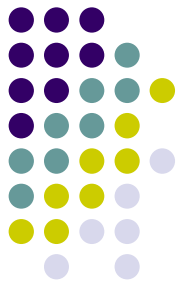
- Source compositions constant
- Chemical species add linearly
- All contributing sources included
- Source profiles linearly independent
- Number of sources less than number of species
- Measurement uncertainties random, uncorrelated, and normally distributed

CMB Model Test Methods and Results

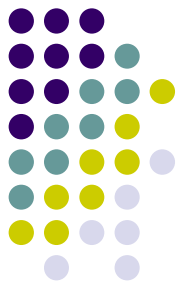


- Constant compositions (substitute different profiles, randomly perturb profiles)– can tolerate substantial variability
- Non-reactive species (estimate profile fractionation, use)– little known
- All source types identified (look for deficits in marker species, unusual values > LQL)– minor contributors can be left out
- Number of sources less than number of species (always the case)– the larger the difference the better

CMB Model Test Methods and Results



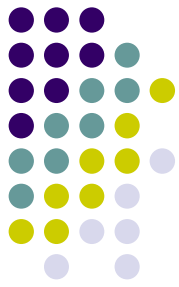
- Source contributions linearly independent (MPIN, collinearity clusters)– degree depends on variability of source profile
- Measurement error distribution (randomized tests with non-normal distributions)– non much difference



5. Adjust Model Inputs

- Increase uncertainties of profile abundances or provide different profile composites
- Create “aged” source profiles with aerosol evolution model
- Identify and characterize missing sources.
- Measure additional species at source and receptor. Stratify samples by meteorological regime

6. Verify Consistency and Stability



- Substitute different profiles for the same source type
- Add or drop species from the fit
- Examine source contributions to species.
- Examine modified psuedo inverse matrix

7. Evaluate and Reconcile Source Apportionments

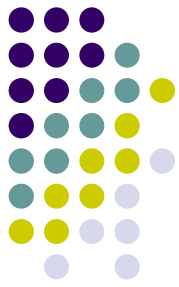


- Compare source contributions among nearby sites
- Compare source contribution variations over time with expected emissions and meteorological variations.
- Apply other receptor methods and compare results
- Apply dispersion models and compare results

Protocol for Reconciling Differences Among Receptor and Dispersion Models



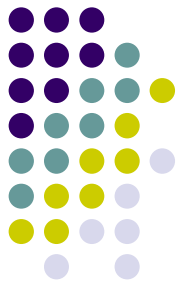
1. Compare CMB and DM results.
2. Verify input data in both models.
3. Recompare results.
4. Refine CMB model inputs.
5. Recompare results.
6. Refine dispersion model inputs.
7. Recompare.
8. "... if it is clearly evident that the dispersion model is not valid, the CMB estimates should be used as the basis for control strategy development. However, if the disparity is not clearly attributable to either model alone, the dispersion model should be used for control strategy development."



Conclusions

- The applications and validation protocol results in more accurate source apportionments.
- Though the protocol does not solve every problem encountered in the CMB, it does identify that a problem exists and suggests some alternatives for solving it.
- Reconciliation of CMB source apportionments with other source apportionment methods yields more accurate results.

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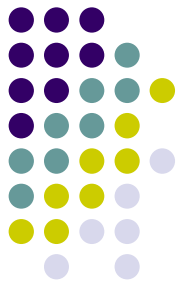
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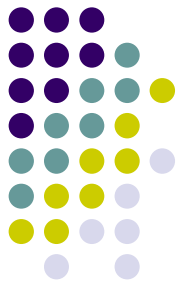
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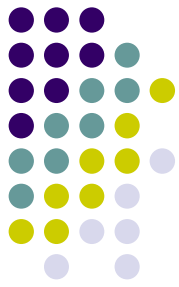
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