Receptor Model Source Apportionment for Air Quality Management

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Department of Physics, University of Genoa, and
L' Istituto Nazionale di Fisica Nucleare (INFN)
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Objectives

- Provide an overview of receptor-oriented source apportionment methods applicable to $PM_{2.5}$
- Provide examples of common pitfalls, limitations, and uncertainties in source apportionment studies and how to overcome them
- Recommend practical source and receptor measurements that are needed to integrated into long term PM networks

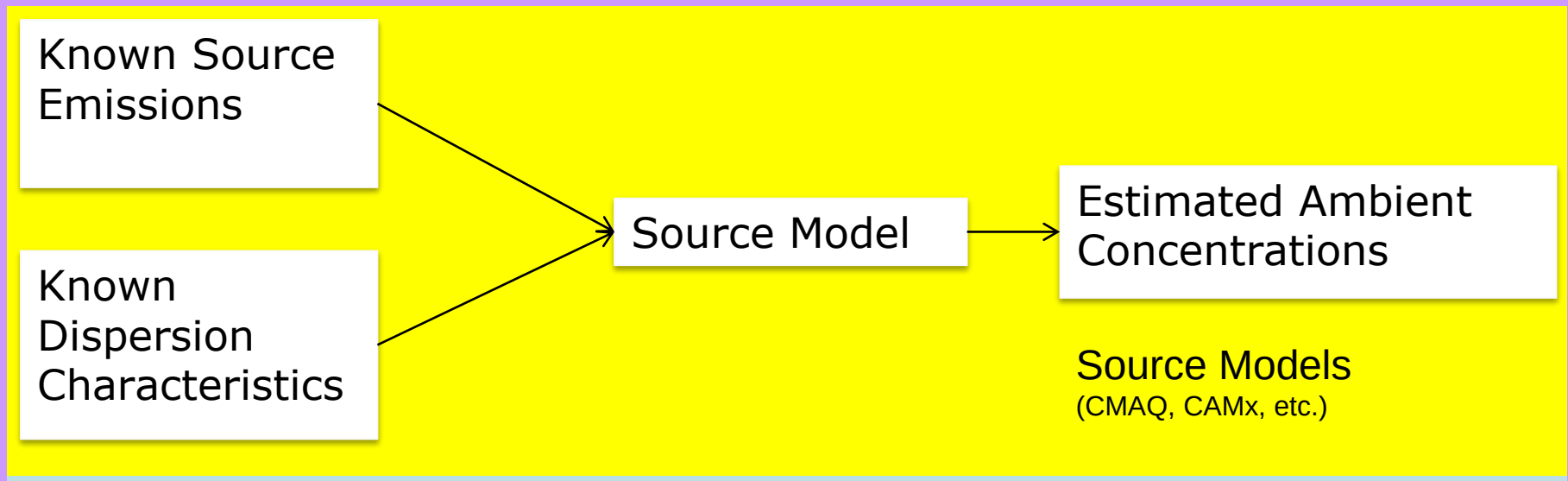
Receptor models have made large contributions to air quality management for 40 years

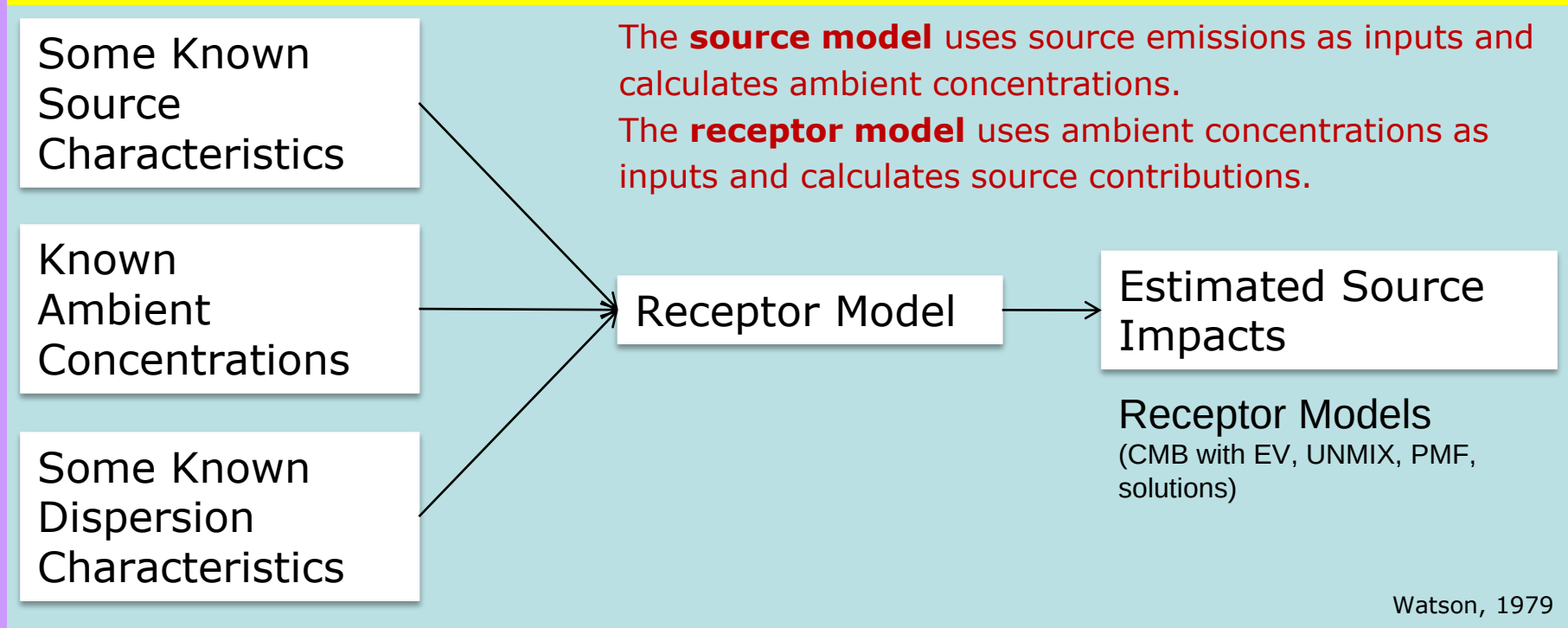
- Identified uninventoried sources as important contributors (wood combustion, cooking, biogenics, road dust, secondary organic and inorganic aerosol, high emitters)
- Focused emission inventory improvements (addition of new categories, refined emission factors for old ones
- Separated primary emittants from secondary formation products (sulfates and nitrates)
- Allowed development of conceptual models for interactions among emissions, meteorology, chemical transformations, and ambient concentrations
- Still the only method to estimate contributions from intermittent and fugitive emissions

There are many U.S. examples of receptor model results being used to formulate emission reduction policies

- 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 (NH_3) regulation (SCAQMD, 1996)

Receptor models are complementary with, not replacements for, source models





The **source model** uses source emissions as inputs and calculates ambient concentrations.

The **receptor model** uses ambient concentrations as inputs and calculates source contributions.

Watson, 1979

The future holds several challenges for receptor modeling

- Pollution controls have eliminated many of the elemental markers
- Secondary organic aerosol has become a larger portion as primary emissions decrease
- Common availability of modeling software and speciated data sets has led to publication of many spurious results

Source and receptor models derive from the same physical construct

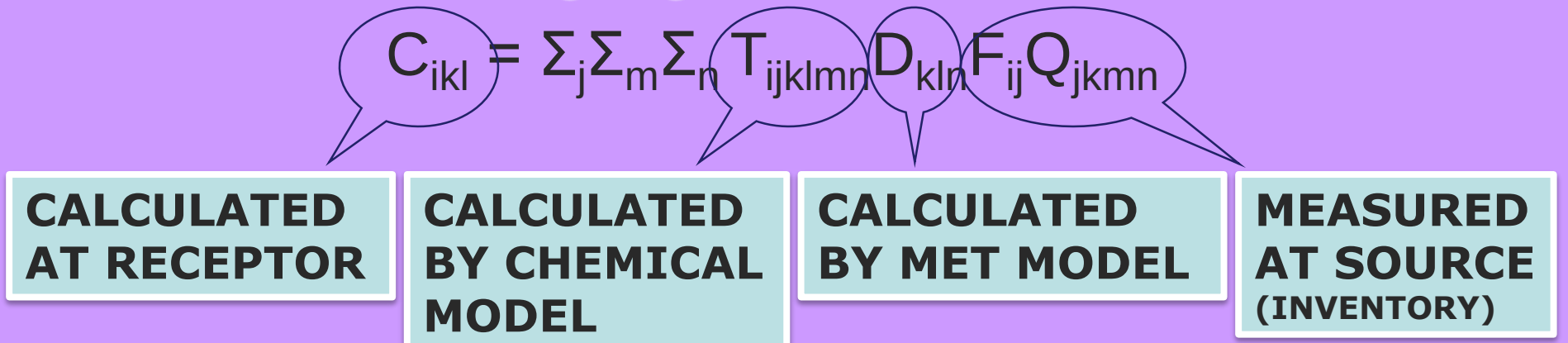
$$C_{ikl} = \Sigma_j \Sigma_m \Sigma_n F_{ij} T_{ijklmn} D_{kln} Q_{jkmn}$$

i	=	pollutant
j	=	source type
k	=	time period
l	=	receptor location
m	=	source sub-type, a specific source or groups of emitters with similar source compositions and/or locations
n	=	location of emitter m of source type j
C_{ikl}	**=**	**ambient concentration**
F_{ij}	=	fractional quantity of pollutant i in source j
T_{ijkmn}	=	transformation of pollutant i during transport
D_{kln}	=	dispersion and mixing between source and receptor
Q_{jkmn}	=	emissions rate

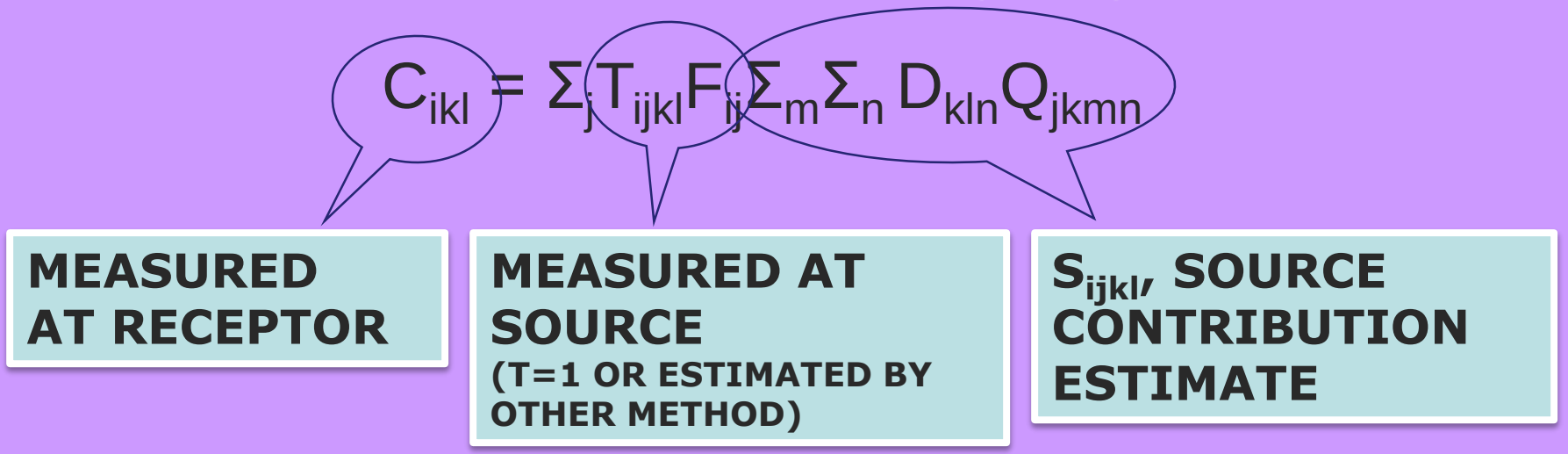
*applies to a specific size fraction for PM

Source and receptor models are complementary with, not replacements for each other

Lagrangian Source Model



Chemical Mass Balance (CMB) Model



Four common solutions are applied to infer source contribution estimates from the CMB equations

Tracer (TR) solution

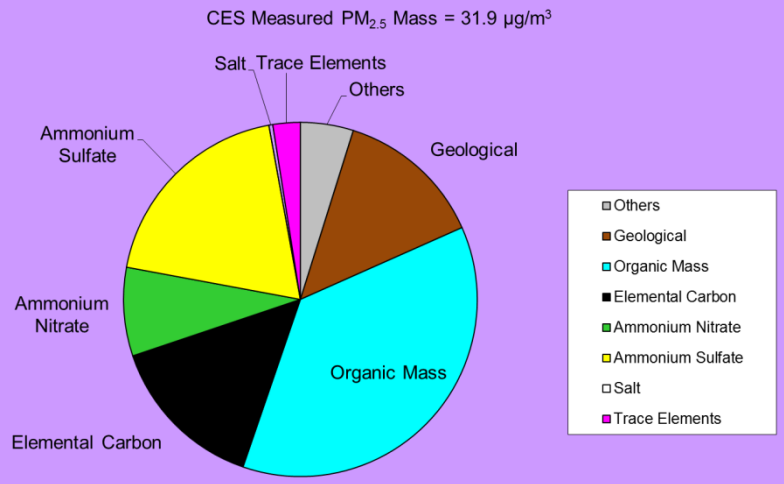
TR-CMB: $S_j = C_i / F_{ij}$

- Assumes the tracer (marker) comes only from, or is dominant in, a specific source type
- "Diagnostic ratio" is a variation that looks at relative abundances of two markers
- Carbon-14 is most common tracer to separate biogenic from fossil carbon sources
- If a marker is available and is not quantified, source identification is not justified

Four common solutions are applied to infer source contribution estimates from the CMB equations

Reconstructed Mass (RM) solution

(RM = Inorganic Ions + OM + EC + Minerals + Salt + Trace Elements + Others)



- Secondary inorganic ions
  (i.e., sum of $SO_4^=$, NO_3^-, and NH_4^+ or sum of $(NH_4)_2SO_4$ and NH_4NO_3)
- Organic mass (OM)
  (i.e., OM=f×OC; f=1.2–1.8)
- Elemental carbon
- Geological minerals (e.g., oxides of Al, Si, Ca, Ti, Fe, and sometimes K)
- Salt (e.g., Na^++Cl^-, 1.4486Na+Cl, or 1.8 Cl^-)
- Trace elements (e.g., sum of remaining measured species excluding double counting)
- Others (e.g., remaining mass, particle-bound water, non-crustal K (K–0.6Fe)=1.2(K–1.6Fe); H_2O, and non $SO_4^=$ S)

Four common solutions are applied to infer source contribution estimates from the CMB equations

Effective Variance (EV) solution

EV-CMB: $\varkappa^2 = \min \Sigma_i [(C_i - \Sigma_j F_{ij} S_j)^2 / (\sigma_{Ci}^2 + \Sigma_j \sigma_{Fij}^2 S_j^2)]$

- Explicitly incorporates uncertainty estimates in both ambient measurements and source profiles
- Positive biases in one abundance are offset by negative biases in others

Four common solutions are applied to infer source contribution estimates from the CMB equations

Positive Matrix Factorization (PMF) solution

$$\text{PMF-CMB: } \varkappa^2 = \min \Sigma_i \Sigma_k \left[(C_{ik} - \Sigma_j F_{ij} S_{jk})^2 / \sigma_{Cik}^2 \right]$$

- Operates on time series from same location, multiple locations, or multiple times and locations
- Restricts all values to non-negative numbers
- Assumption that source profile measurements is untrue

Deviations from CMB assumptions must be evaluated for all solutions

- Compositions of source emissions as perceived at the receptor are constant over the period of ambient and source sampling.
- Chemical species from different sources do not react with each other.
- All sources with a potential for significant contribution to the receptor have been identified and have had their emissions characterized.
- The source compositions are linearly independent of each other.
- Measurement errors are random, uncorrelated, and normally distributed.
- The number of source types is less than or equal to the number of chemical species.

CMB applications and validation protocols need to be adapted to address these new challenges and to accommodate all of the solution methods

United States Environmental Protection Agency | Office of Air Quality Planning and Standards Research Triangle Park NC 27711 | EPA-450/4-87-010 May 1987

Air

EPA

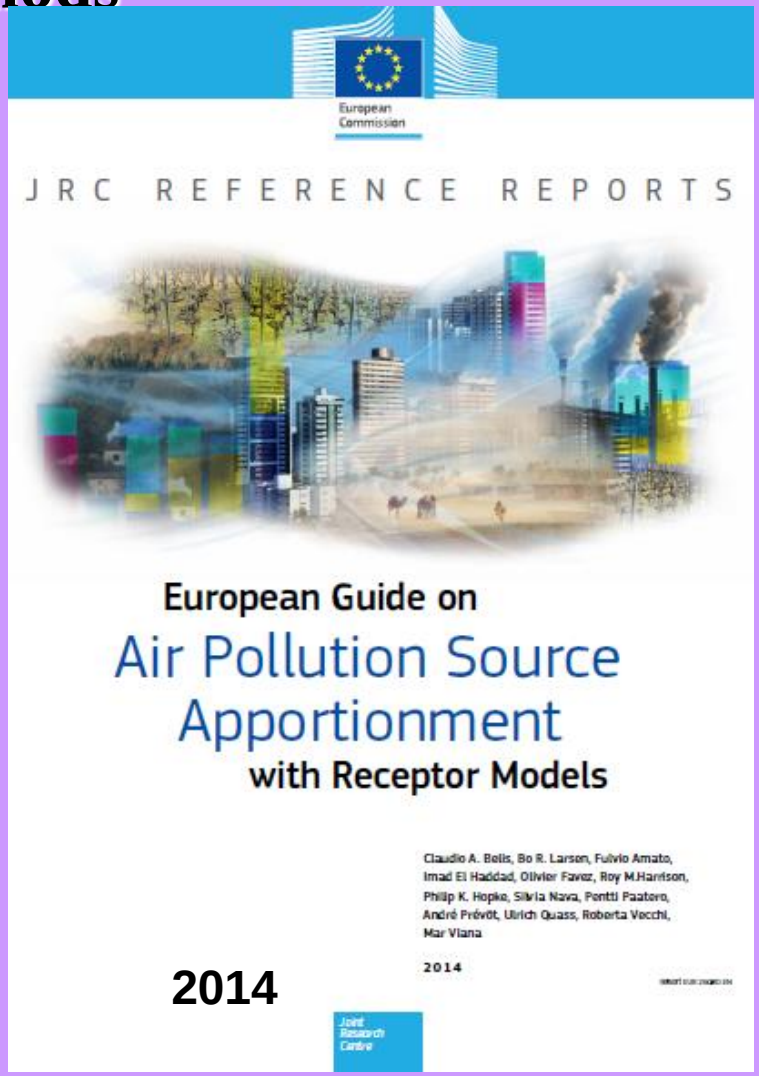
Protocol For Applying And Validating The CMB Model

1987

Finalized in 1998, but dated 2004

Protocol for Applying and Validating the CMB Model for $PM_{2.5}$ and VOC

www.epa.gov/scram001/models/receptor/CMB_Protocol.pdf



2014

http://source-apportionment.jrc.ec.europa.eu/Docu/EU_guide_on_SA.pdf

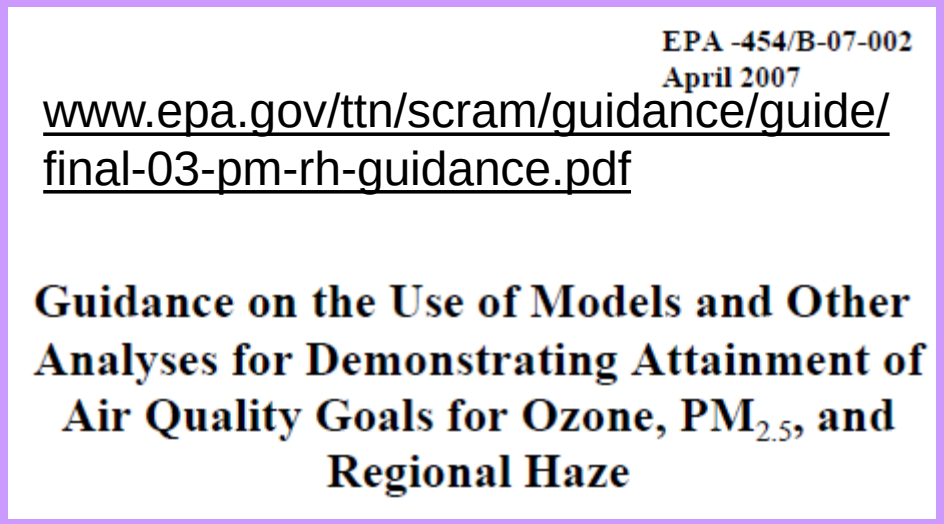
Complex mathematics are not needed to identify the major source categories or quantify their contributions

- Tracer solution identifies source types
- Reconstructed mass solution classifies and quantifies major source categories
- Spatial, temporal, and PM size patterns make source obvious
- Nearly all elevated levels will contain: 1) secondary sulfates and nitrates; 2) engine exhaust; and 3) fugitive dust.
- Some will contain biomass burning and salt
- Specific sources require additional marker species

Guidance specifies extensive validation and evaluation protocols, but these are rarely followed

1. Determine model and solution applicability
2. Format input files and perform initial source apportionment
3. Evaluate outputs and performance measures
4. Evaluate deviations from model assumptions
5. Modify model inputs to remediate problems
6. Evaluate the consistency and stability of the model results
7. Corroborate CMB results with other modeling and analyses

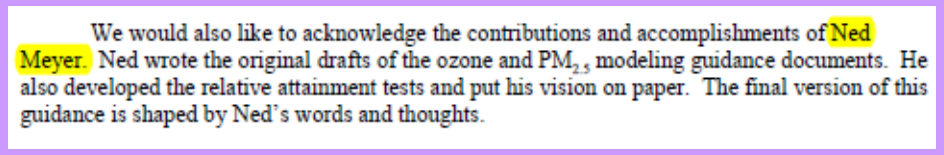
Step 7 has been further elucidated as a "weight of evidence" evaluation



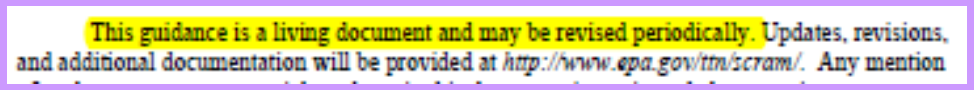
EPA -454/B-07-002
April 2007

www.epa.gov/ttn/scram/guidance/guide/final-03-pm-rh-guidance.pdf

Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze



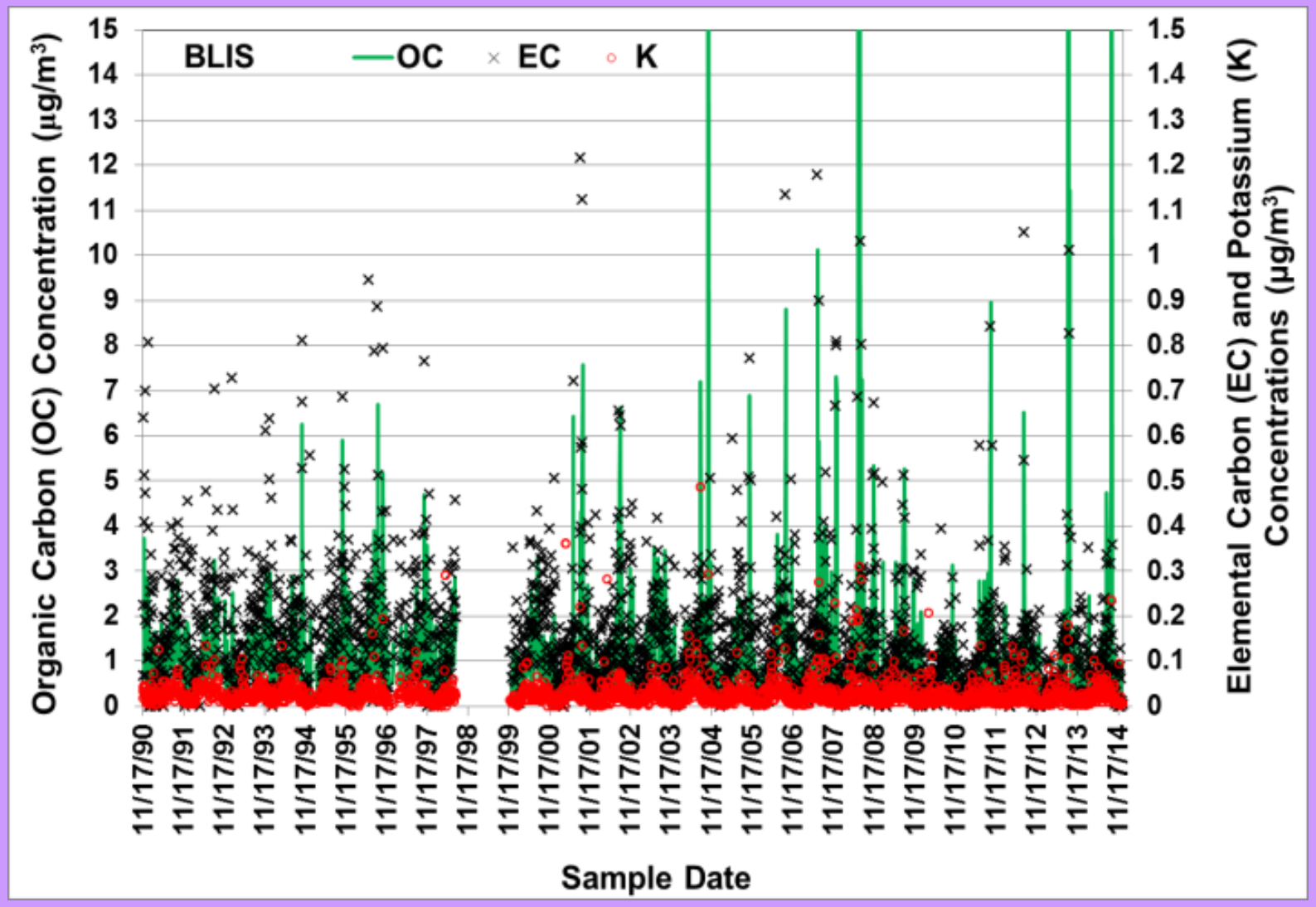
We would also like to acknowledge the contributions and accomplishments of Ned Meyer. Ned wrote the original drafts of the ozone and $PM_{2.5}$ modeling guidance documents. He also developed the relative attainment tests and put his vision on paper. The final version of this guidance is shaped by Ned's words and thoughts.



This guidance is a living document and may be revised periodically. Updates, revisions, and additional documentation will be provided at *http://www.epa.gov/ttn/scram/*. Any mention

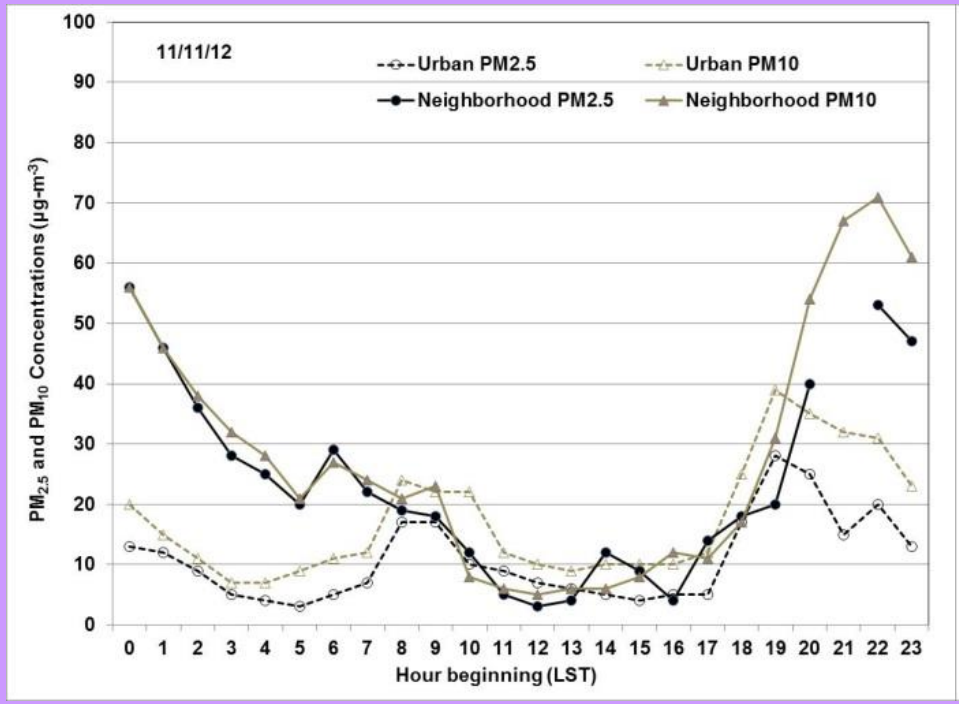
- Examine the problem using different methods
- Use discrepancies between model results to identify and correct weaknesses in models and input data
- Quantify confidence intervals
- Explain and qualify conclusions regarding source contribution estimates

Wildfires are obvious cause of excursions at non-urban Lake Tahoe

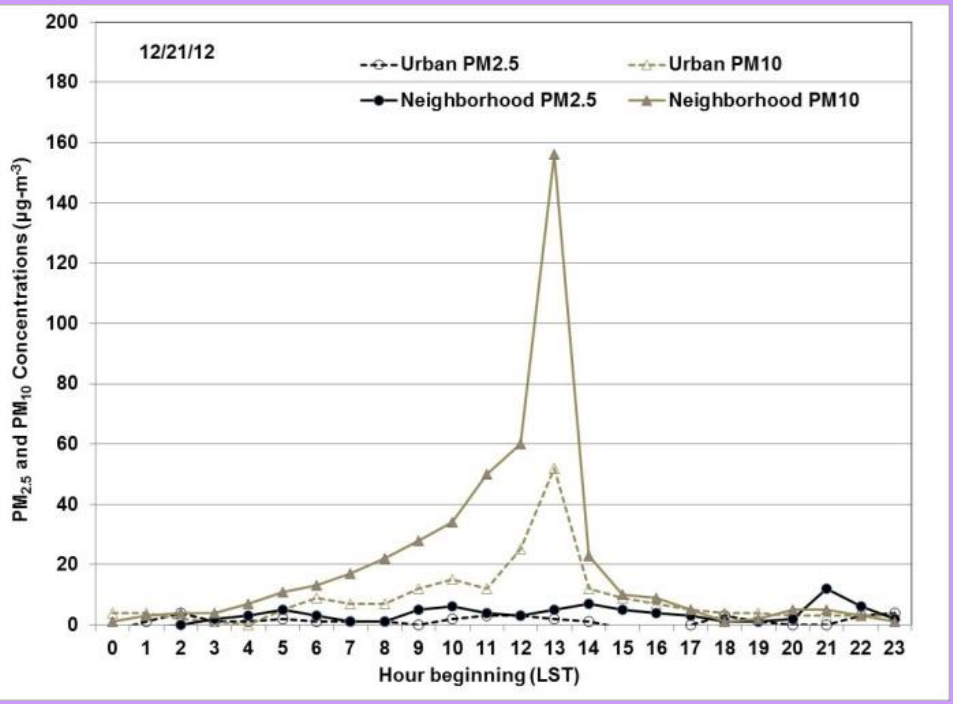


Temporal and spatial patterns indicate sources and magnitudes

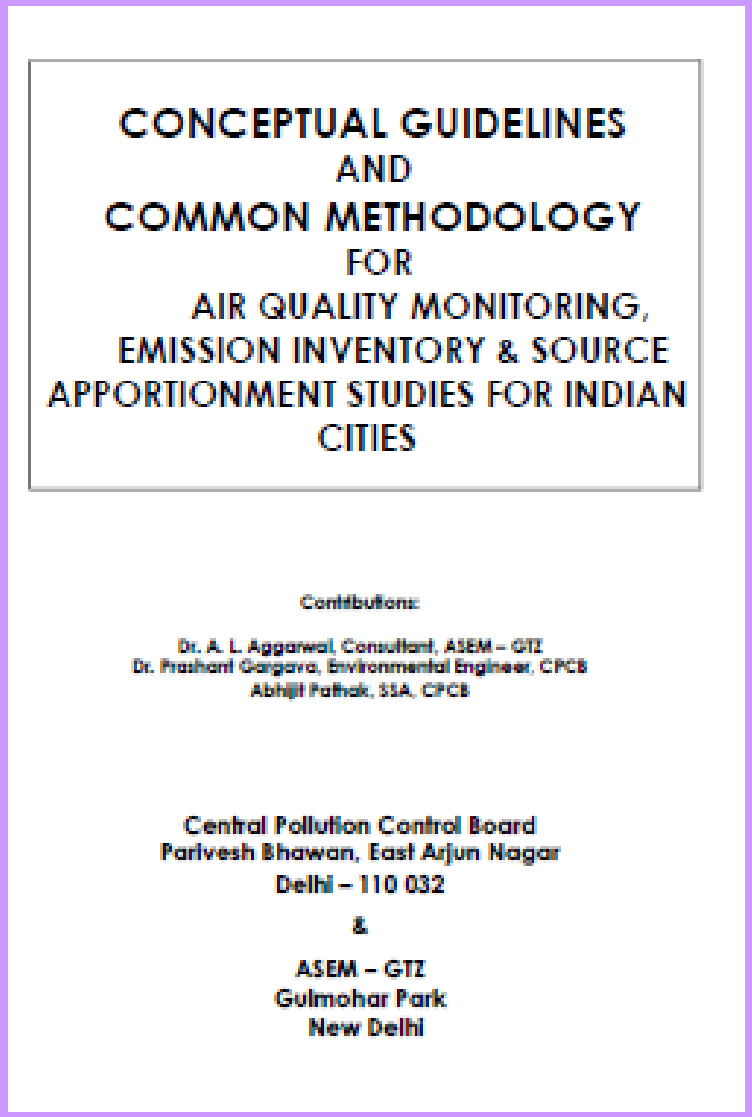
Wintertime Wood Smoke



Fugitive Dust from De-icing



Weight of evidence was lacking in a recent EV-CMB source apportionment in India



CONCEPTUAL GUIDELINES
AND
COMMON METHODOLOGY
FOR
AIR QUALITY MONITORING,
EMISSION INVENTORY & SOURCE
APPORTIONMENT STUDIES FOR INDIAN
CITIES

Contributions:

Dr. A. L. Aggarwal, Consultant, ASEM – GTZ
Dr. Prashant Gargava, Environmental Engineer, CPCB
Abhijit Pathak, SSA, CPCB

Central Pollution Control Board
Parivesh Bhawan, East Arjun Nagar
Delhi – 110 032

&

ASEM – GTZ
Gulmohar Park
New Delhi

- Good start
  - Network was well designed
  - Source types were identified and characterized
  - Marker species were measured
  - Source and receptor species were compatible

Danger of Ignoring the Weight of Evidence:

LPG most polluting? Experts disagree

Chetan Chauhan
chetan@hindustantimes.com

NEW DELHI: A government claim that the source of the Capital's deadliest pollutant Particulate Matter 2.5 is liquid petroleum gas (LPG) in homes and not vehicles has miffed experts who term it as an attempt to give the transport sector a clean chit for air pollution.

PM 2.5, the smallest pollutant absorbed mostly by the human body, can trigger heart attacks and respiratory diseases.

Rise in number of vehicles was believed to be the major source of the pollutant.

This claim was countered by Indian Oil Corporation this week when it quoted a Central Pollution Control Board study saying LPG was the major contributor to rising PM 2.5 in the Capital.

EXPERTS SAY GOVT GIVING TRANSPORT SECTOR CLEAN CHIT FOR POLLUTION WITH STUDY SHOWING LPG AS BIGGEST POLLUTANT

An IOC presentation at a seminar organised by diesel vehicle manufacturers said that half of PM 2.5 in residential areas of Delhi was because of combustion of domestic LPG. In industrial areas, it was as high as 61 per cent and at traffic junctions 40.5 per cent.

"It is not a complete view," said CPCB chairperson S.P. Gautam. The board for the first time in India conducted an air pollution source appropriation study which was peer reviewed by air pollution experts from Europe and the US and is being examined by an inter-ministerial group. "I don't know what IOC had said but there are many factors which contribute to particulate matter."

The most intriguing findings were for residential areas in Delhi where vehicles contribute 22.4 per cent and kerosene combustion 17.4 per cent to total PM 2.5 pollution.

The presentation states vehicles contribute only seven per cent to particulate matter at traffic intersections and garbage burning for 14 per cent.

"It is shocking," said Anumita Roy Chowdhury, Associate Director with NGO Centre for Science and Environment. "Refinery and auto industries have hyped data in public forums to prove vehicles are the cleanest and must be left alone."

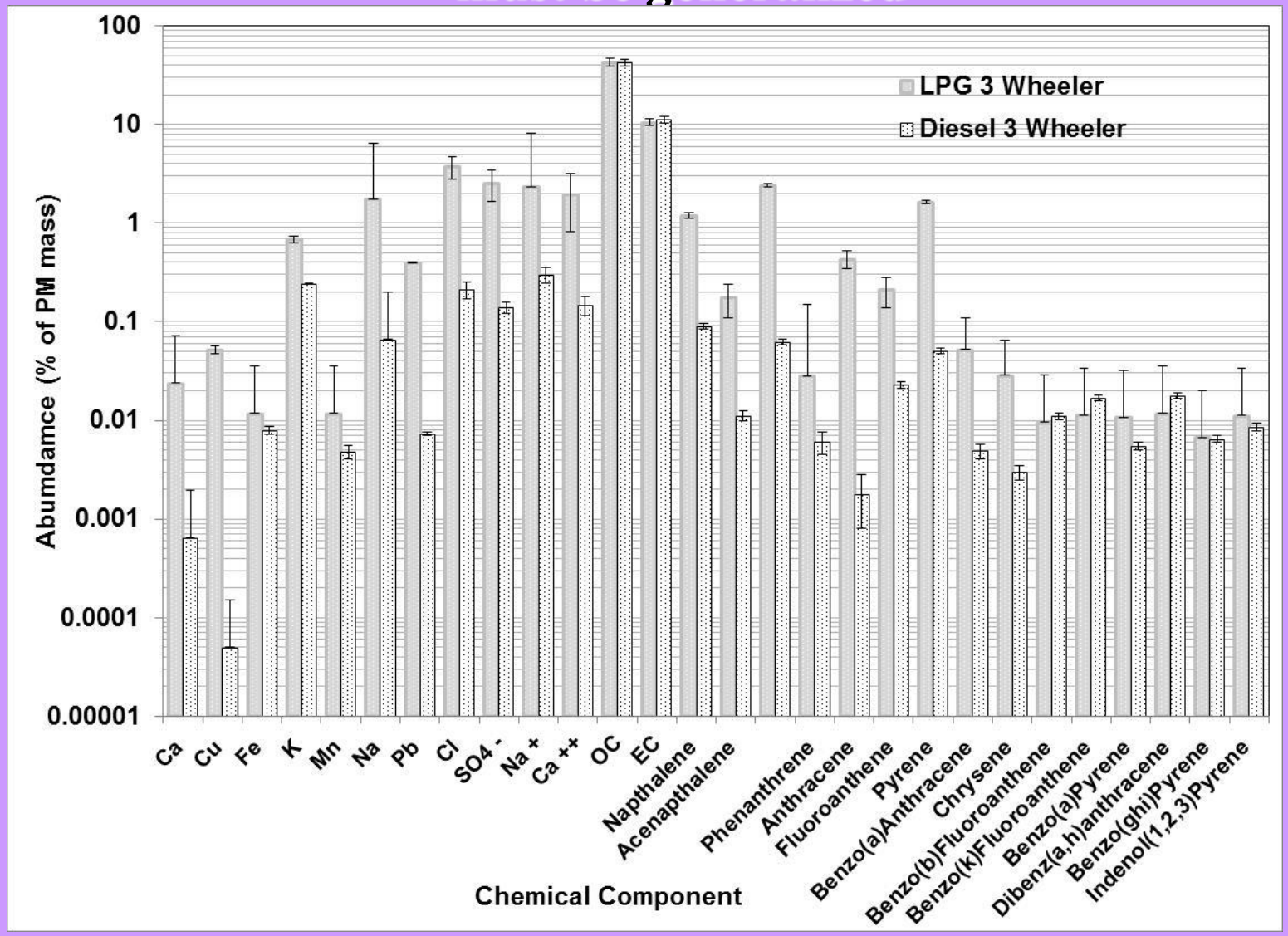
The CPCB study, which Environment Minister Jairam Ramesh has decided not to put in public domain, is likely to be the basis for India's future auto fuel policy. The government has constituted an inter-ministerial group to review the present policy, which expires in 2010, and create one for the new decade.

Environment ministry officials said the aim of the new policy would be to reduce the sources of air pollution.

Chowdhury said the government was framing a new policy without consulting people.

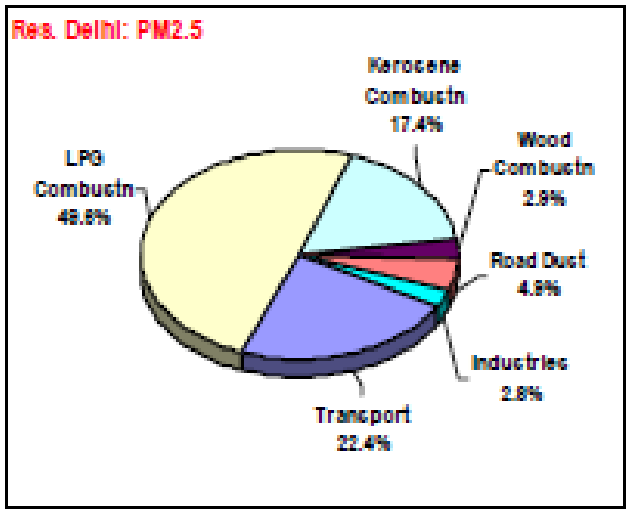
- Weight of evidence would include external data from vehicle and stove emission tests, comparisons with apportionments from different cities, examination of other data such as continuous gas and particle measurements.

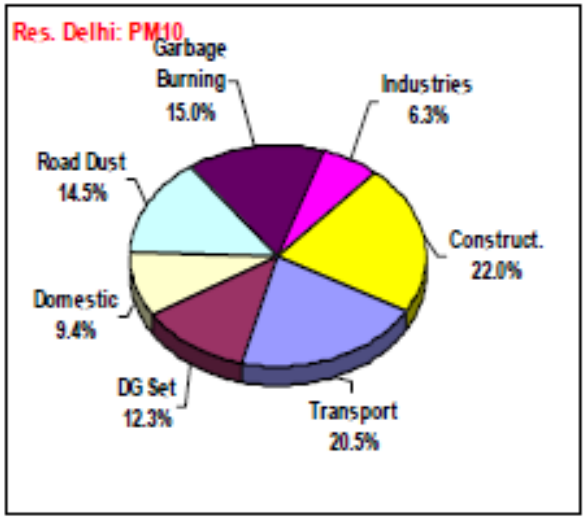
Sensitivity tests would have shown that the measured LPG and diesel exhaust profiles are collinear, and their source designation must be generalized

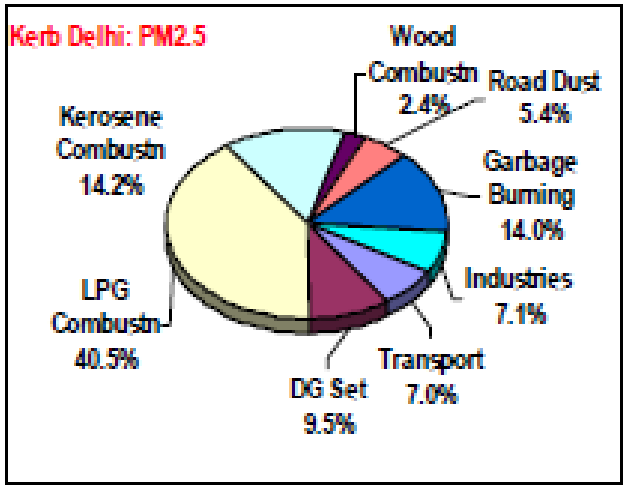


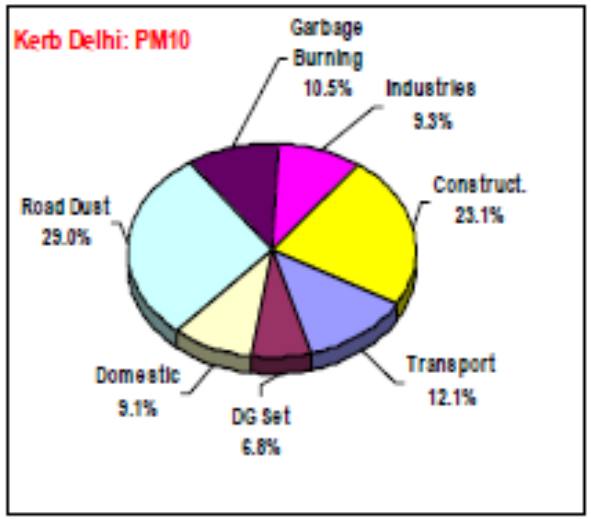
Sethi and Patil, 2008a,b

Internal consistency tests would have revealed discrepancies between size fractions and sampling locations





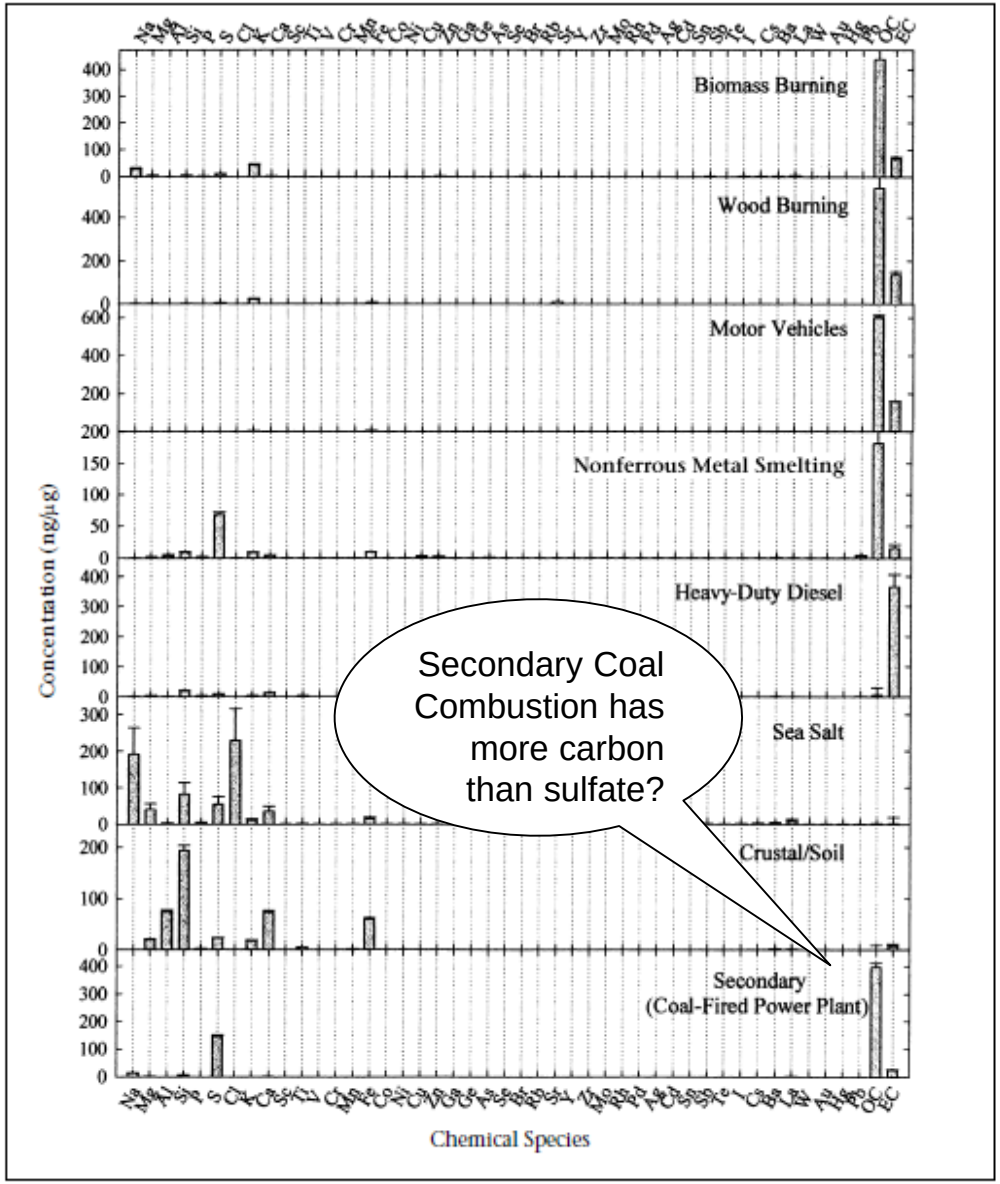




NEERI, 2010

Chemical source profiles derived from ambient data don't always make sense if not from a source-dominated environment

- PMF (Positive matrix factorization)-derived factors from ambient data often have mixing of different sources, yet they are identified as a single source profile

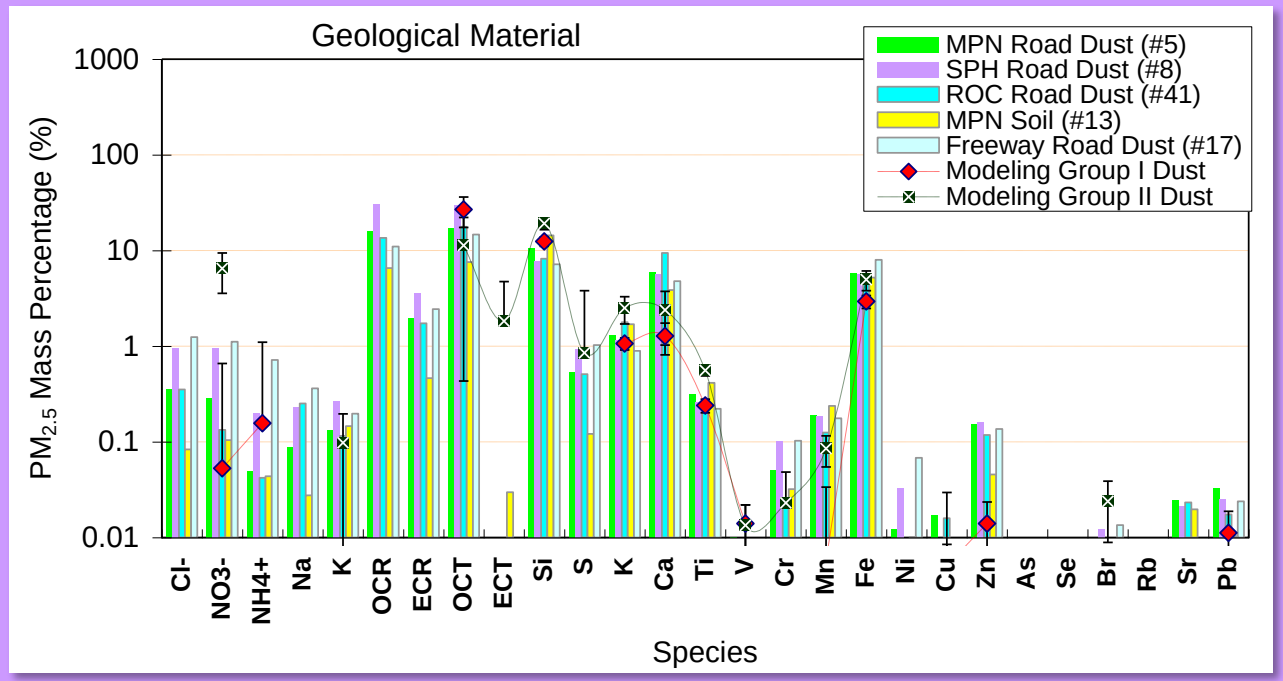


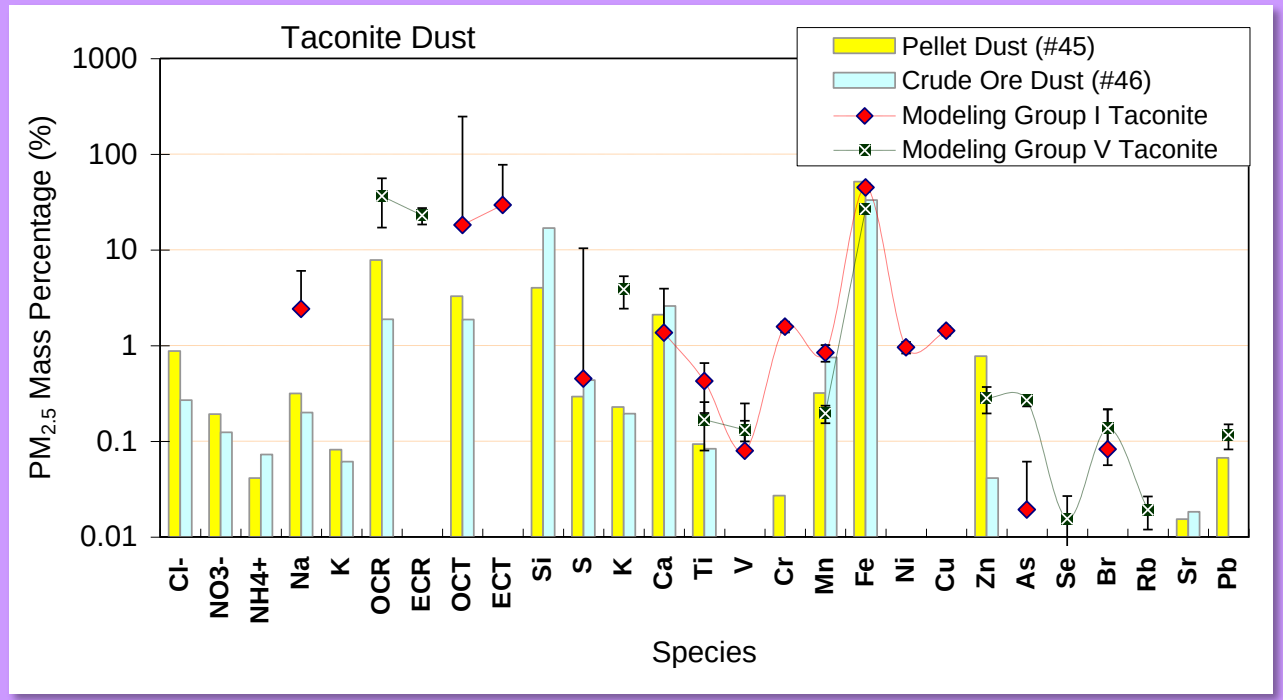
Ramadan et al. (2000) , *JAWMA*, p. 1308-1320

Figure 1. Source profiles for DFPSS fine particle samples with Q = 35,067 and FPEAK = −0.20.

Each PMF and Unmix source factor should be compared with at least one measured profile

Example from Minnesota

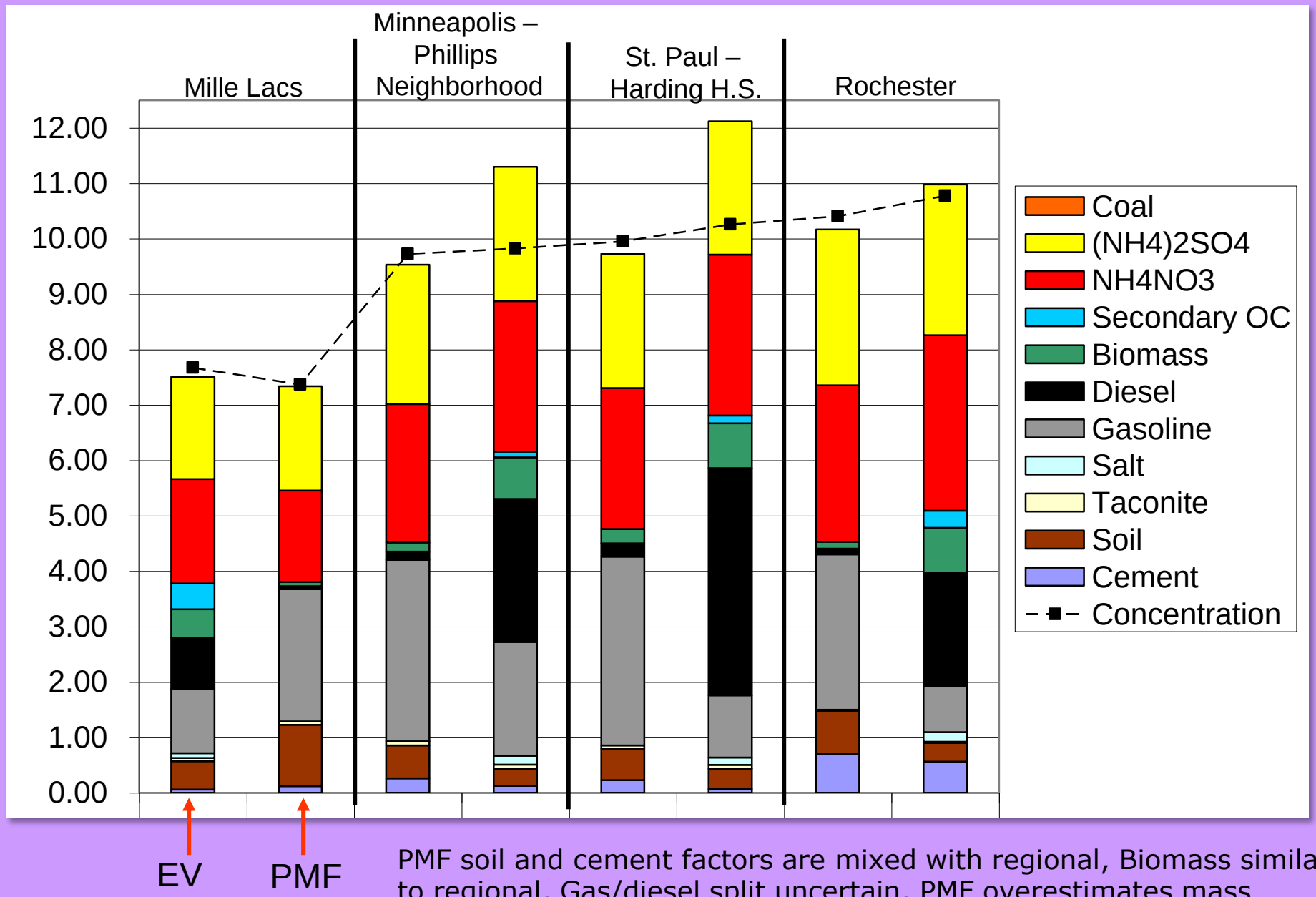




Chen et al. 2010, 2011

Applying different CMB solutions to the same data set aids in the Weight of Evidence

(Minnesota, 8/2003 – 7/2004, most samples passed validation tests)

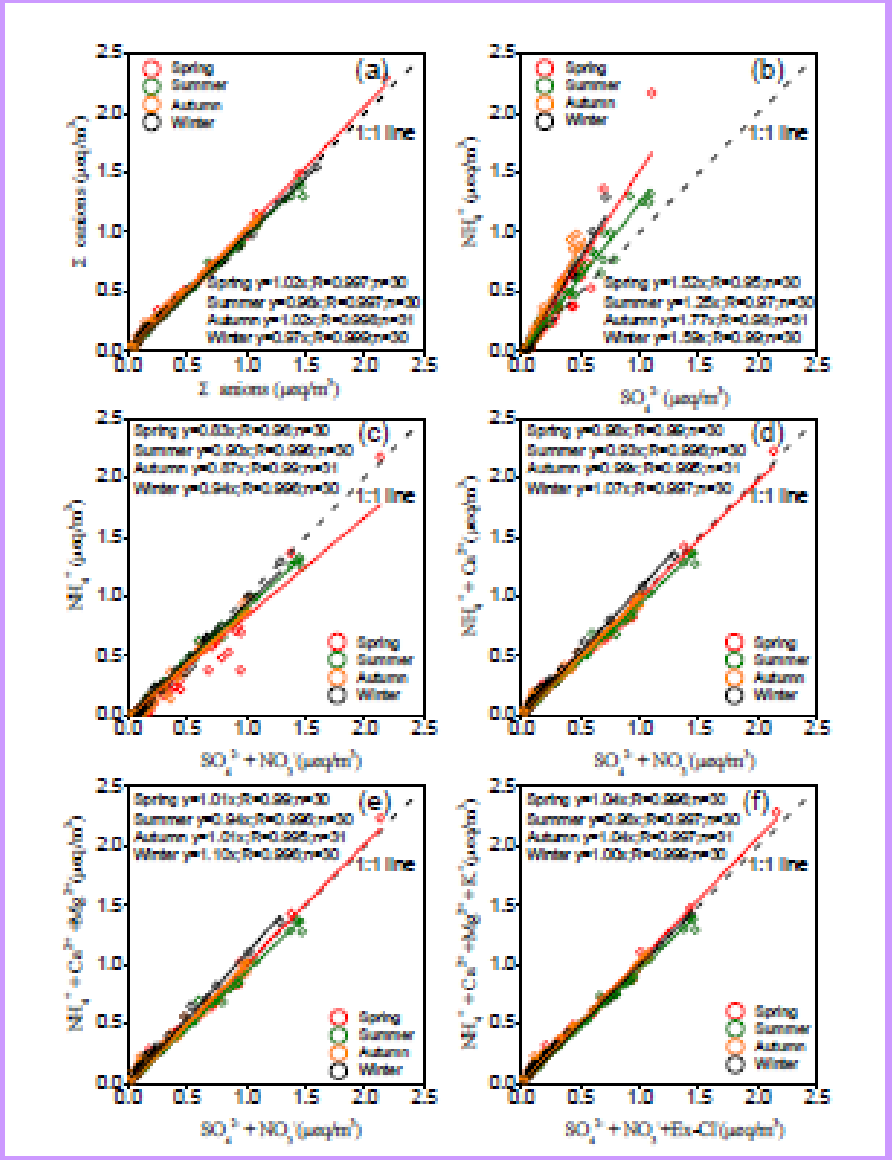


PMF soil and cement factors are mixed with regional, Biomass similar to regional, Gas/diesel split uncertain, PMF overestimates mass

Chen et al., 2011

Data validation for $PM_{2.5}$ source apportionment at Peking University was good

SO_4 was totally neutralized by NH_4, indicating that contributions are more regional than local

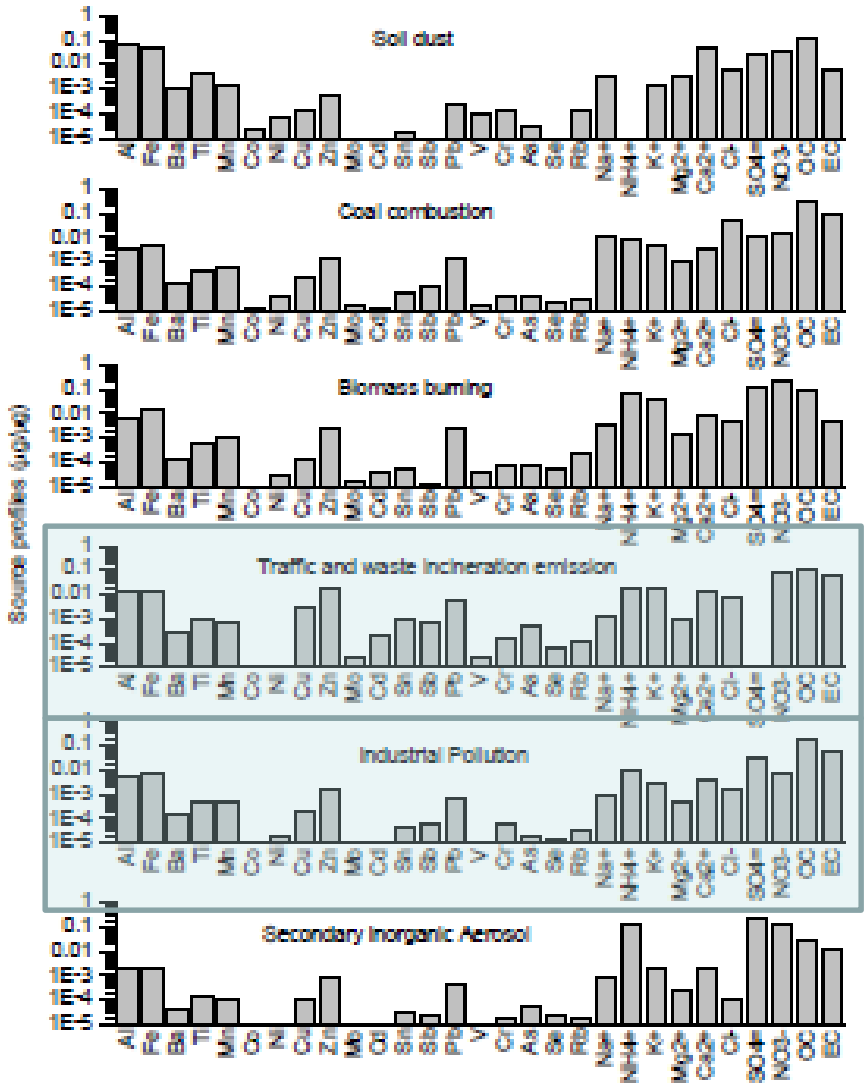


Zhang, et al., 2013

But the "Industrial Pollution" factor at road-centric Peking University doesn't make sense!

Table 2. Relative contributions from six identified sources of $PM_{2.5}$ in Beijing within the one-year and four-season periods.

Source	Spring	Summer	Autumn	Winter	Annual
Soil dust	23%	3%	18%	16%	15%
Coal combustion	5%	1%	7%	57%	18%
Biomass burning	19%	6%	17%	7%	12%
Traffic and waste incineration emission	5%	4%	4%	2%	4%
Industrial pollution	14%	52%	42%	12%	25%
SIA	34%	34%	13%	6%	26%



PMF analysis of elements, ions, and carbon at PKU 4/2009 to 1/2010

Zhang, R.; Jing, J.; Tao, J.; Hsu, S.C.; Wang, G.; Cao, J.J.; Lee, C.S.L.; Zhu, L.; Chen, Z.; Zhao, Y.; Shen, Z. (2013). Chemical characterization and source apportionment of PM2.5 in Beijing: seasonal perspective. *Atmos. Chem. Phys.*, **13**(14):7053-7074. http://www.atmos-chem-phys.net/13/7053/2013/.

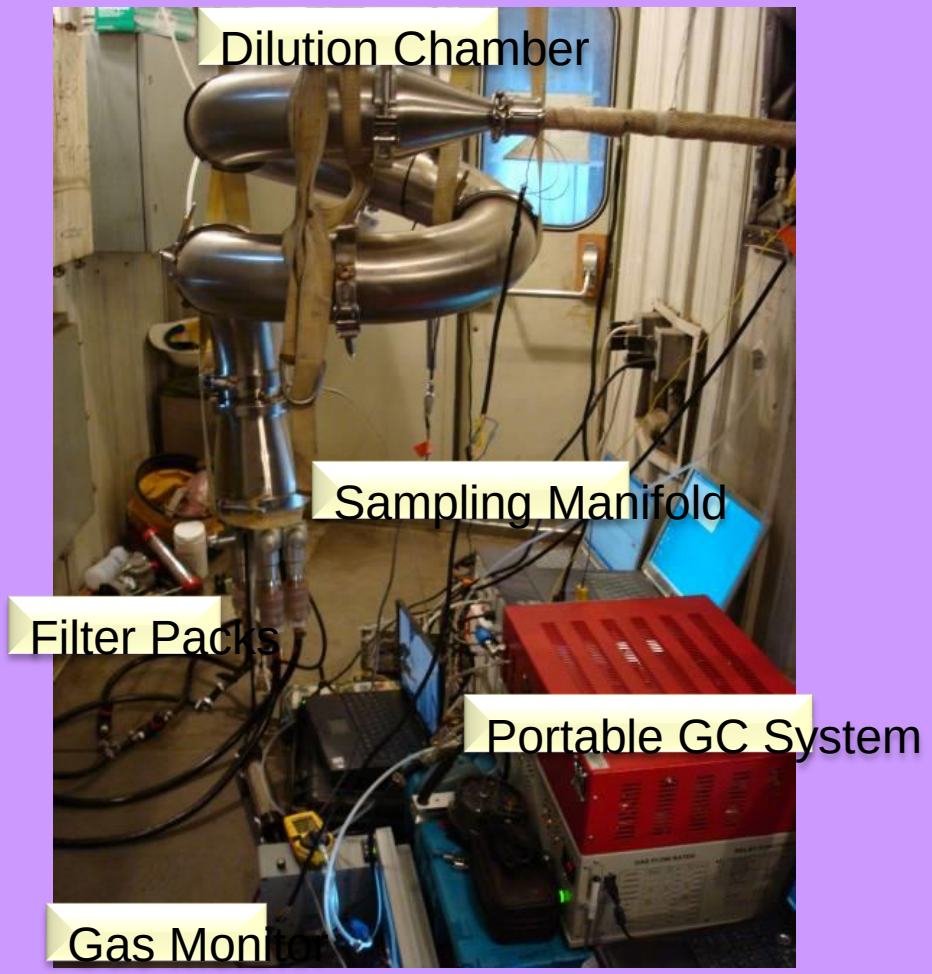
Major limitation of receptor modeling is lack of evolution for modern source and receptor measurements

- Source compliance tests do not represent real-world emissions
- PM speciation networks are limited to mass, elements, ions, and carbon fractions
- Source profile and PM speciation data bases are not available, kept up to date, or equivalent

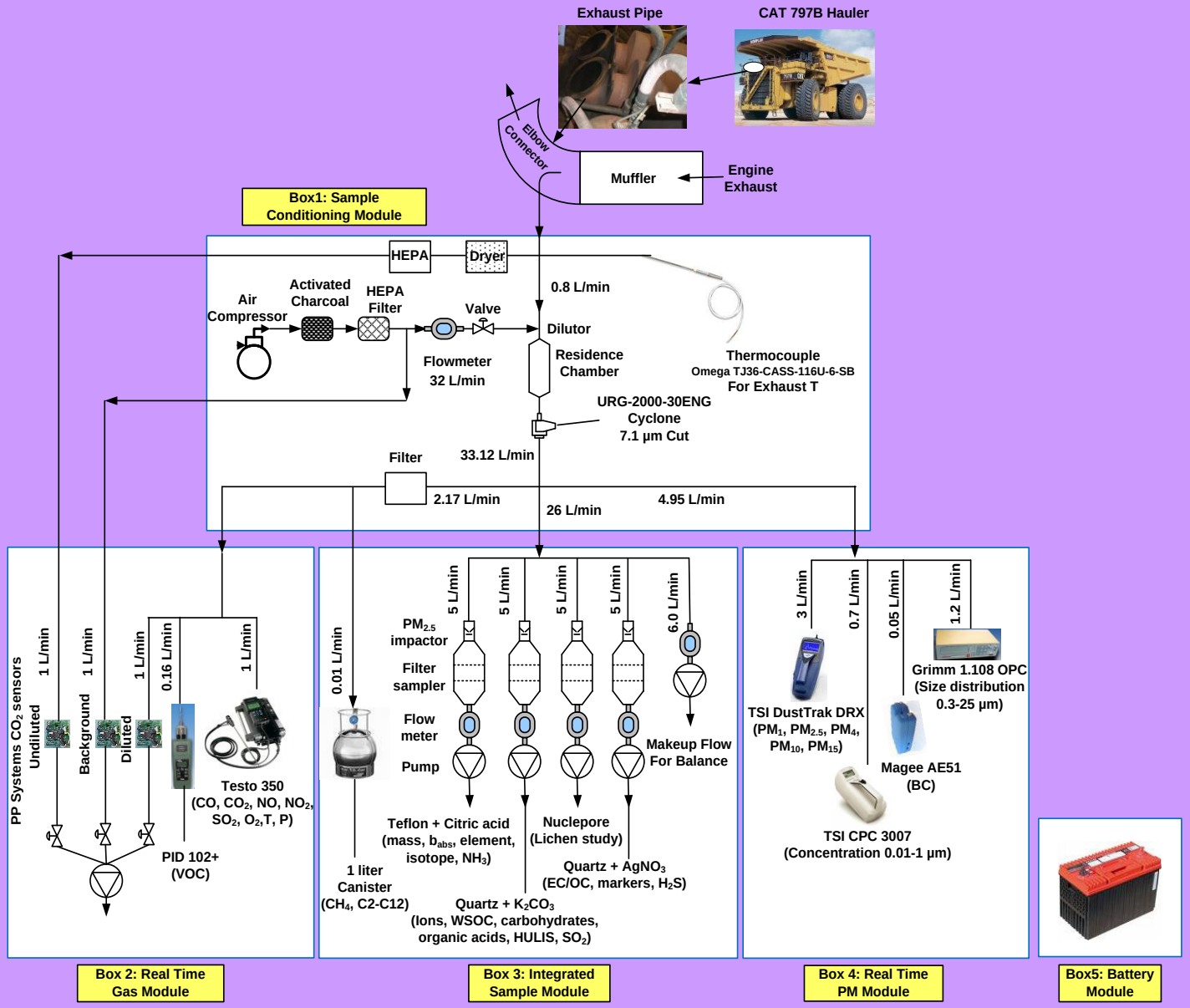
Compliance tests could be made more useful for source apportionment

U.S. EPA's compliance sampling method is obsolete

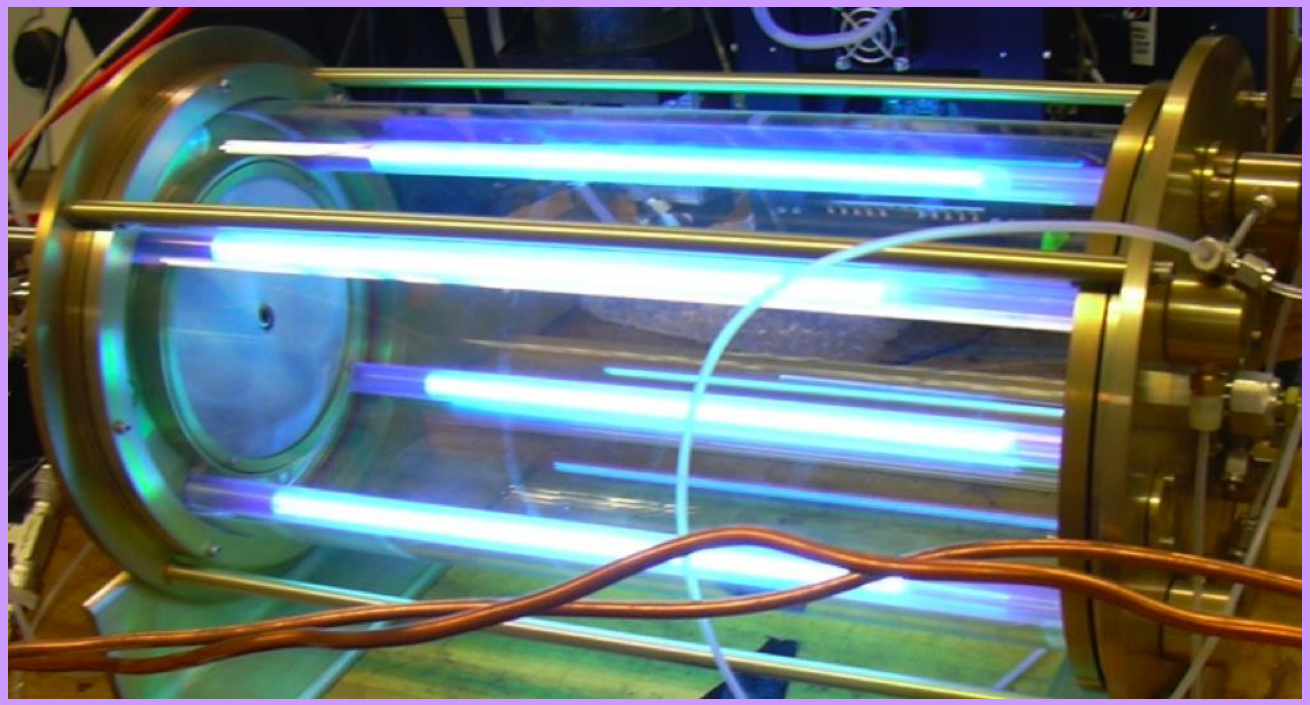
Dilution sampling better simulates profiles at receptors



New technologies can be combined into multipollutant systems to obtain source profiles as well as emission rates

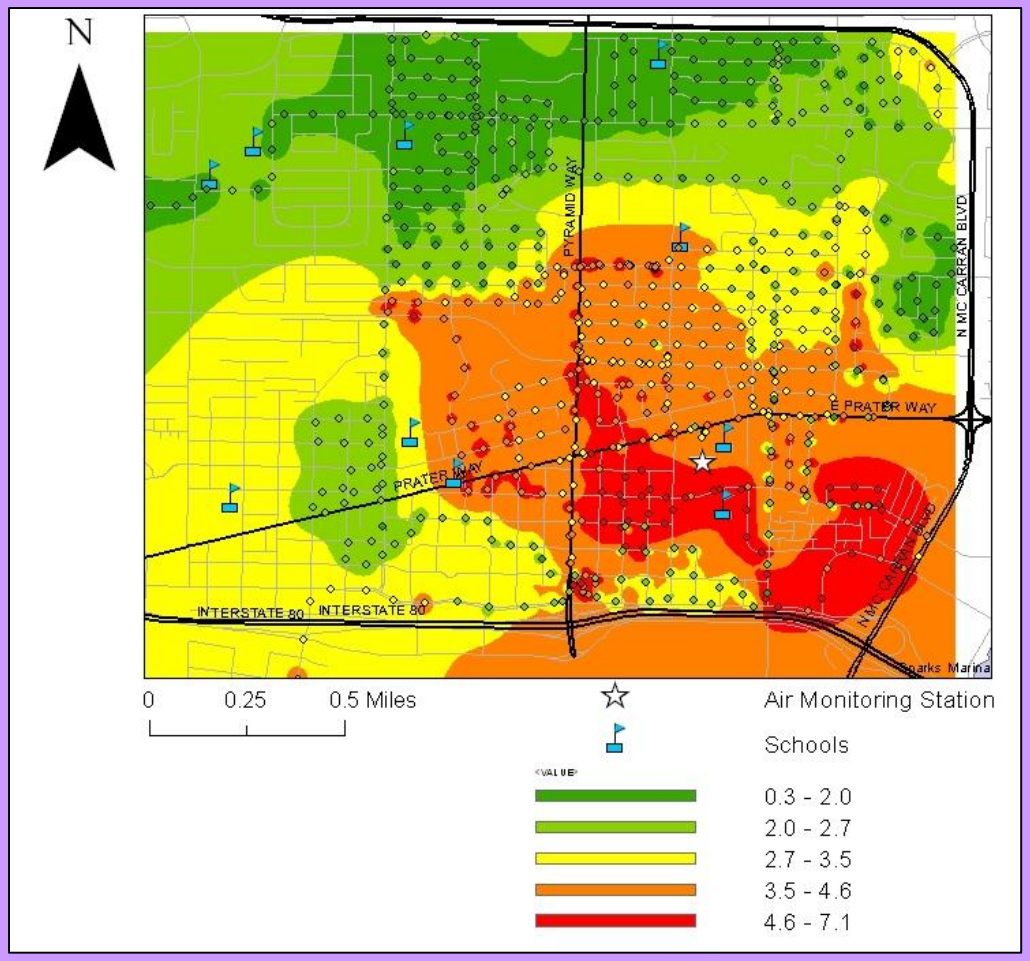


Using a flow tube reactor at the after sampling can simulate atmospheric aging



Microsensor measurements are useful for mapping the zone of influence for source contributions

The brown carbon cloud around a woodburning neighborhood near Reno, Nevada, shows a limited footprint for exposures. Wood stove changeout incentives can be limited to this neighborhood



With gas and particle measurements, $SO_4^{=}/SO_2$ ratio changes during aerosol aging

(and should be reflected in source profiles)

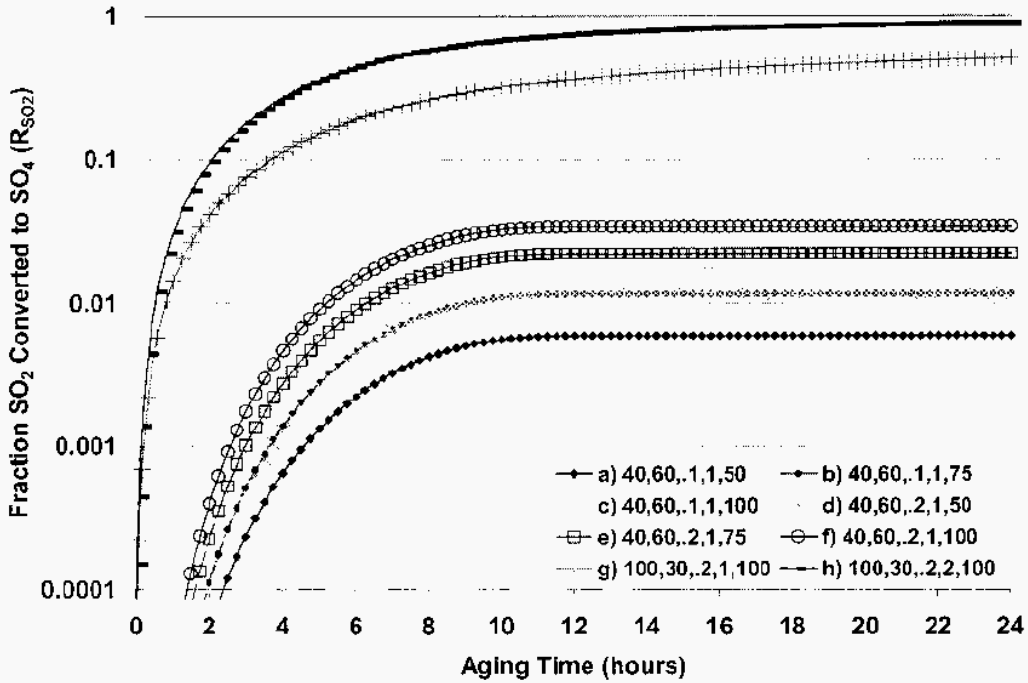


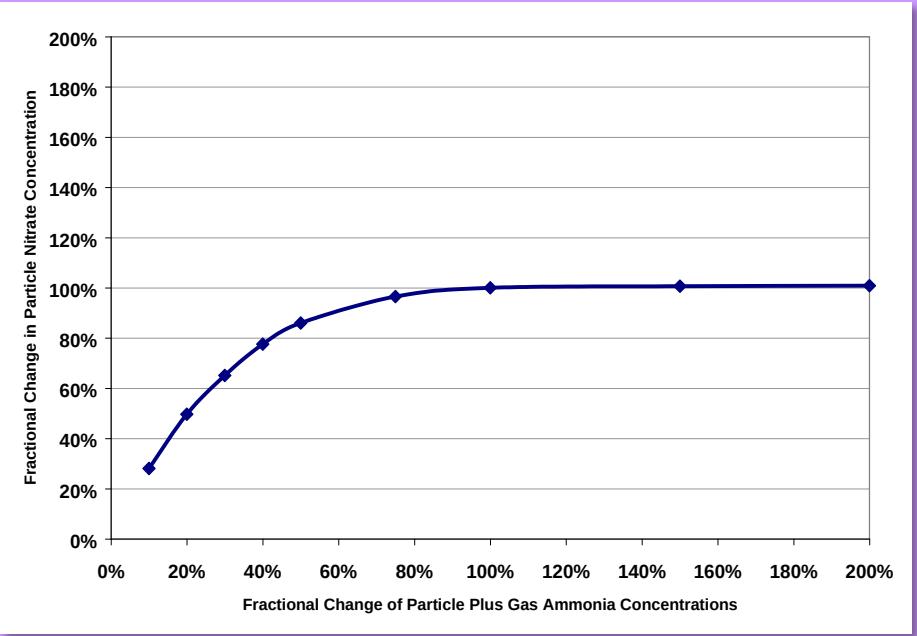
Figure 3. SO_2 transformation fractions from the Aerosol Evolution Model as a function of aging time for different conditions. Sequential numbers identifying each plot are relative humidity (%), ozone (ppbv), formaldehyde (ppbv), hydrogen peroxide (ppbv), and fraction of maximum solar insolation (%).

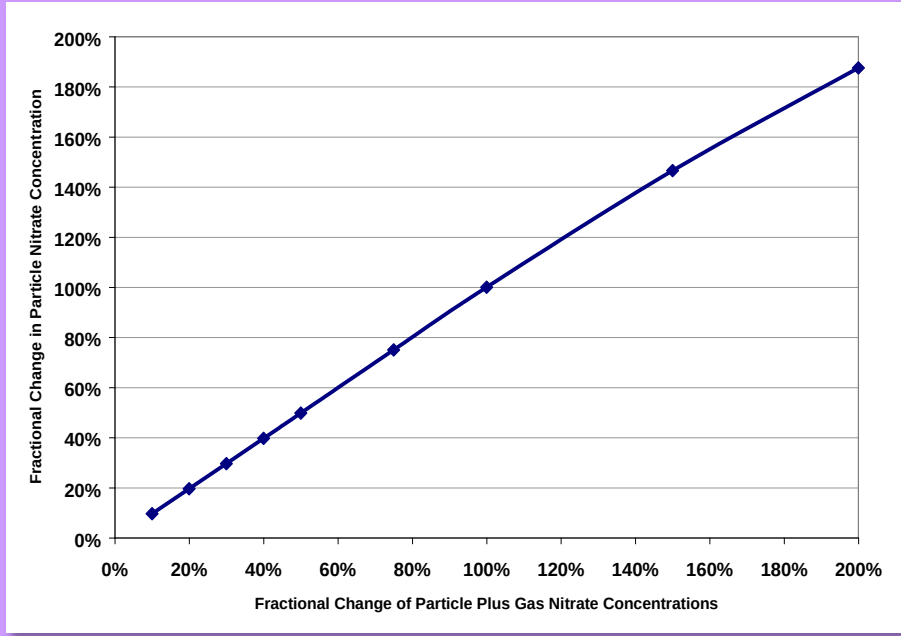
$C_{ikl} = \Sigma_j \Sigma_m \Sigma_n F_{ij} T_{ijklmn} D_{kln} F_{ij} Q_{jkmn}$ (CMB Equation)

T_{ijklmn} = Transformation of pollutant i during transport

Watson et al., 2002, *Energy & Fuels*

Gas and particle measurements can be used to estimate limiting precursors with equilibrium models

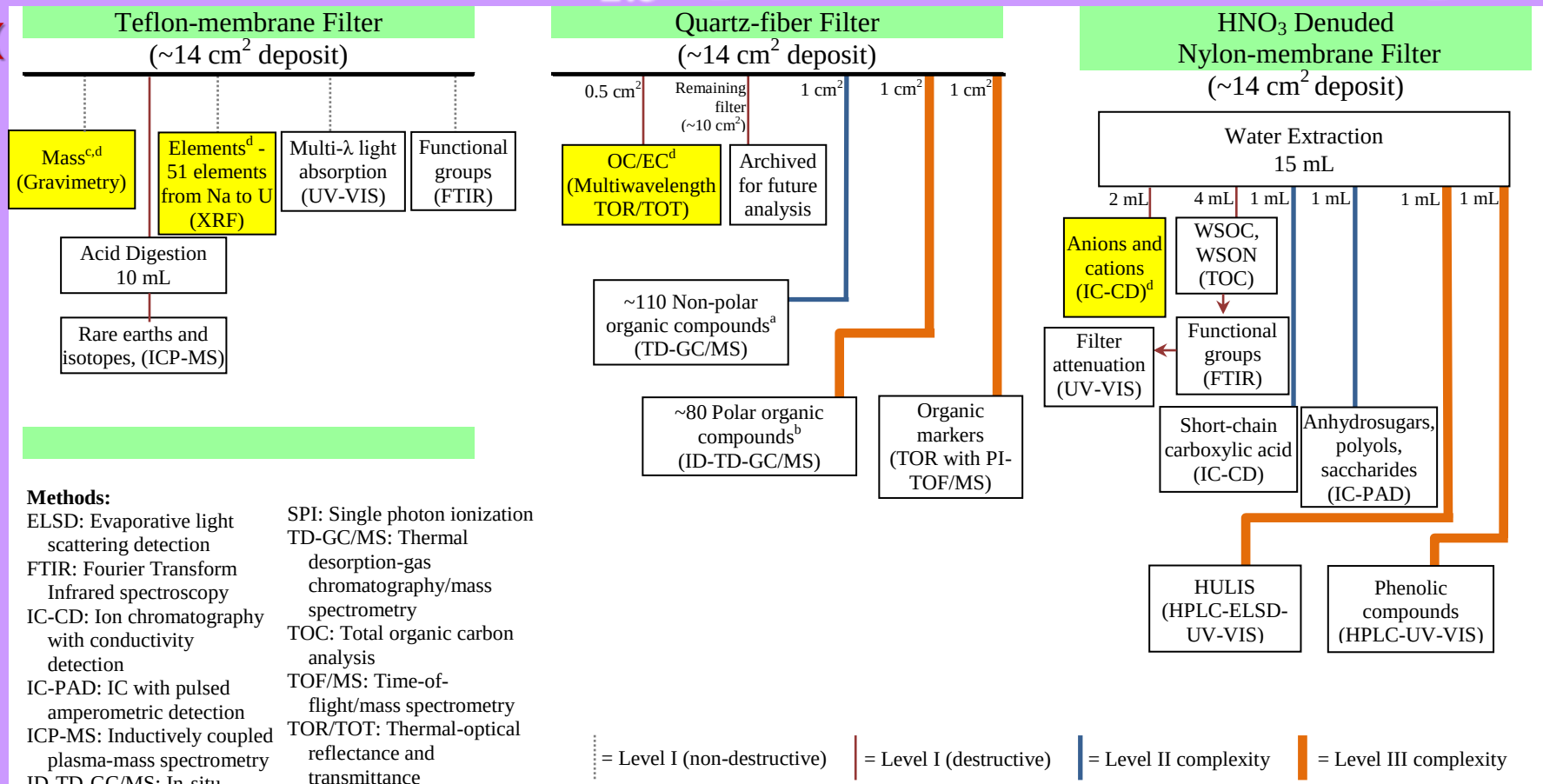




A lot of ammonia needs to be reduced in order to reduce ammonium nitrate concentrations

Nitric acid reduction is more effective to reduce ammonium nitrate concentrations

More source apportionment information can be obtained from $PM_{2.5}$ speciation network samples



Methods:

ELSD: Evaporative light scattering detection
FTIR: Fourier Transform Infrared spectroscopy
IC-CD: Ion chromatography with conductivity detection
IC-PAD: IC with pulsed amperometric detection
ICP-MS: Inductively coupled plasma-mass spectrometry
ID-TD-GC/MS: In-situ-derivatization-thermal desorption-gas chromatography/mass spectrometry
HPLC: High performance liquid chromatography
HULIS: Humic-like substances
SPI: Single photon ionization
TD-GC/MS: Thermal desorption-gas chromatography/mass spectrometry
TOC: Total organic carbon analysis
TOF/MS: Time-of-flight/mass spectrometry
TOR/TOT: Thermal-optical reflectance and transmittance
UV-VIS: Ultraviolet-visible light spectroscopy (200-1100 nm)
XRF: X-ray fluorescence spectroscopy
WSOC: Water-soluble organic carbon
WSON: Water-soluble organic nitrogen

[a] Including alkanes, alkenes, polycyclic aromatic hydrocarbons (PAHs), hopanes, and steranes
[b] Including levoglucosan, other carbohydrates, intermediate and long-chain carboxylic acids (including diacids and multi-functional acids), phenolic compounds, and sterols
[c] Measured in most $PM_{2.5}$ and PM_{10} AAQS compliance networks
[d] Measured in many PM speciation networks

Many new source markers can be obtained from these methods

Major Source Type	Source Sub-Type	Specific Markers
Traffic-related emissions	Road dust	Cu, Fe, Sb, Al, Si, Ba, styrenebutadiene rubber (SBR), benzothiazole (tire wear), asphaltenes (pavement wear)
	Gasoline engine exhaust	17 α(H), 21 β(H)-hopane, 17 α(H)-diastigmastane, other hopanes, steranes, PAH diagnostic ratios
	Diesel engine exhaust	high molecular weight hydroxycarbonyls, hopanes, steranes, PAH diagnostic ratios
Fossil fuel combustion	Fuel oil combustion (e.g., heating oil, kerosene in externally-fired boilers)	PAH diagnostic ratios
	Residual oil combustion (Bunker fuel)	Ni, V
	Uncontrolled coal combustion (e.g., domestic heating and cooking)	Se, As, Cd, Hg, Zn, $SO_4^{=}$, picene, PAH diagnostic ratios
	Controlled coal combustion (e.g., power plants)	Se, As, S, $SO_4^{=}$, picene, PAH diagnostic ratios
Anthropogenic combustion	Tobacco smoke	iso/anteiso alkanes
	Meat cooking	Cholesterol, palmitic acid, palmitoleic acid, stearic acid, oleic acid
	Trash/Plastic burning	1,3,5-triphenylbenzene, tris(2,4-di-tert-butylphenyl)phosphate
Biomass burning	Softwood	resin acids, guaiacol derivatives, retene, levoglucosan/mannosan ratio (3-6)
	Hardwood	syringol derivatives, levoglucosan/mannosan ratio (15-25)
	Straw and grasses	levoglucosan/mannosan ratio (>30)
	Peat	levoglucosan/mannosan ratio (~10)
	Biomass burning SOA	3-methyl-5-nitrocatechol, 3-methyl-6-nitrocatechol, 4-methyl-5-nitrocatechol, 6-nitroguaiacol, 4,6-dinitroguaiacol; HULIS
Natural sources	Mineral dust	Ca, CO_3^{-3}, Si, Al
	Sea salt	Na, Cl
	Bioaerosol – fungi	arabitol, mannitol, ergosterol
	Bioaerosol – bacteria	hydroxy fatty acids
Secondary organic aerosol (SOA)	Anthropogenic SOA	2,3-dihydroxy-4-oxopentanoic acid, o-phthalic acid, 2,4,6-trimethylphenol, oxy-PAHs, oxalic acid, excess OC/EC ratios
	Isoprene-derived biogenic SOA	2-methylthreitol, 2-methylerythritol, 2-methylglyceric acid, cis-2-methyl-1,3,4-trihydroxy-1-butene, 3-methyl-2,3,4-trihydroxy-1-butene, trans-2-methyl-1,3,4-trihydroxy-1-butene, excess OC/EC ratios
	Monoterpene-derived biogenic SOA	cis-pinic acid, cis-pinonic acid, trans-norpinic acid, cis-caric acid, limonic acid, ketolimononic acid, 3-hydroxyglutaric acid, 3-methyl-1,2,3-butanetricarboxylic acid, excess OC/EC ratios

Conclusions

- Receptor model source apportionment has played a positive role in improving air quality management
- Evaluation and validation of source contribution estimates is often neglected and adversely effects air quality management
- Measurement technologies can be updated to better meet the challenges for future source apportionment.

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