The Role of Receptor Models in Creating a Weight-of-Evidence Emission Reduction Strategy:

Analytical and Procedural Approaches

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Desert Research Institute

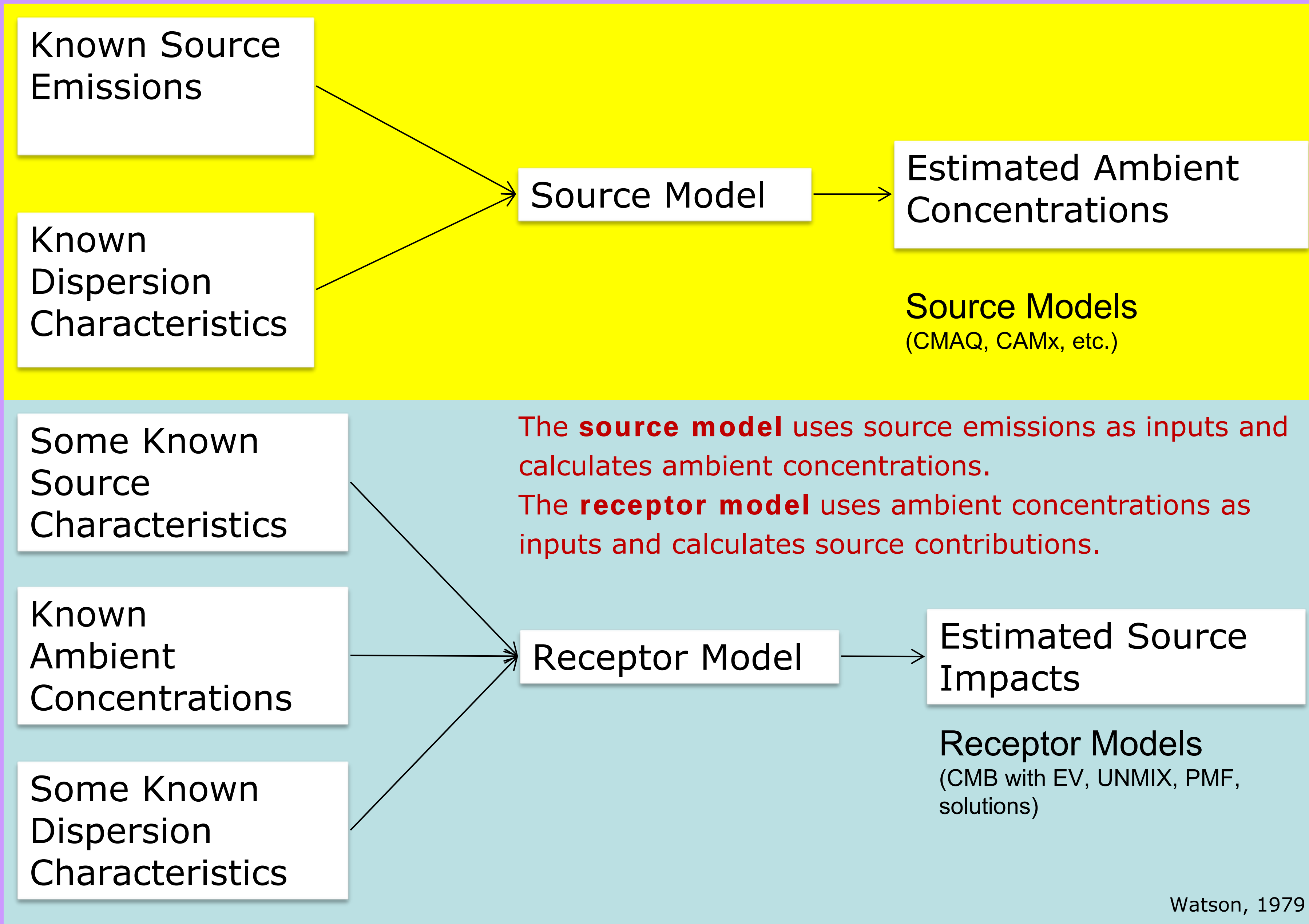
Objectives

- Summarize prior guidance attempts to ensure evaluation and validation of model results
- Provide examples of common pitfalls, limitations, and uncertainties in source apportionment studies and how to overcome them
- Suggest some improvements for receptor model evaluation and evaluation guidance

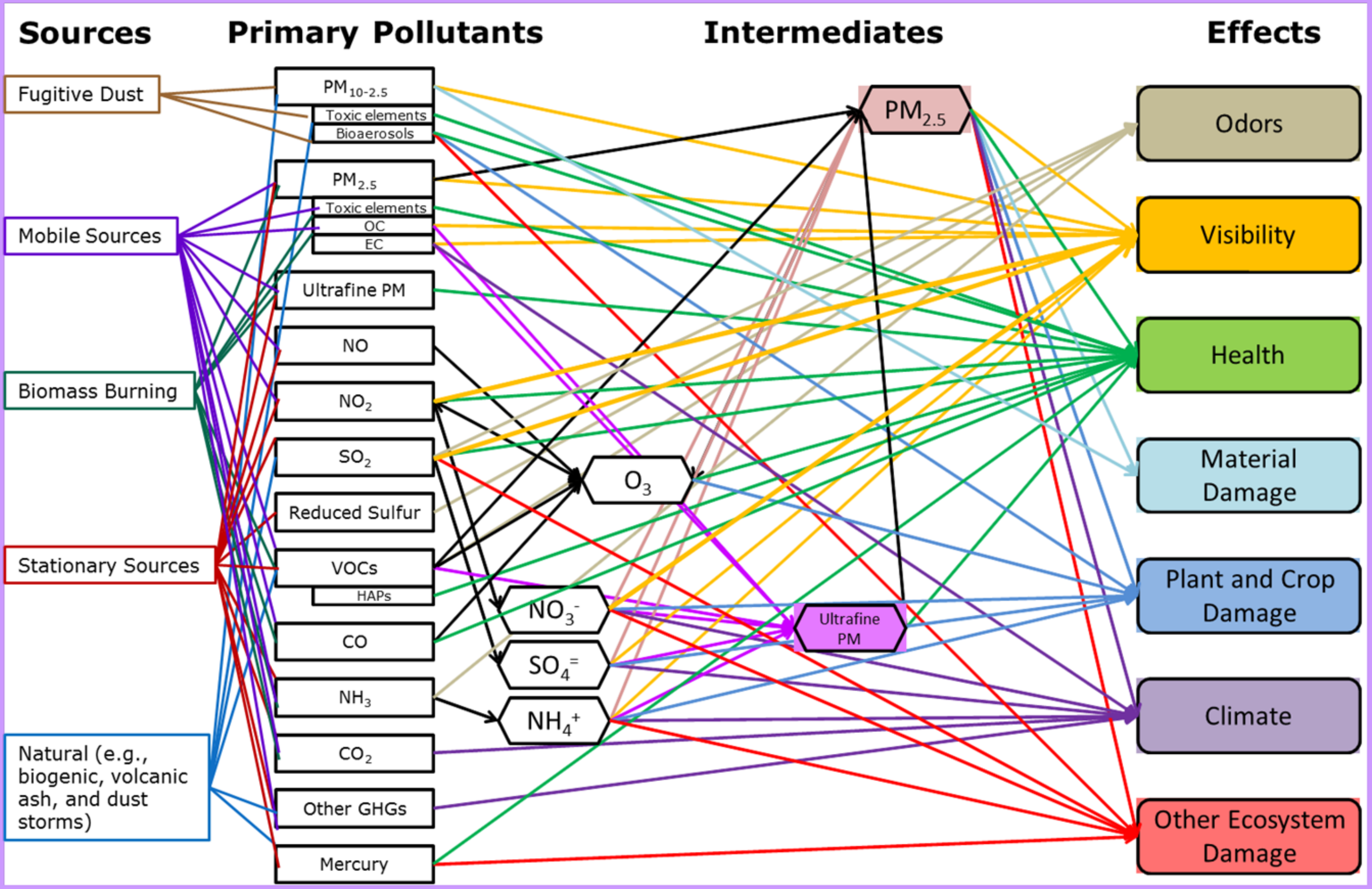
Receptor models have made large contributions to air quality management for 40 years

- Identified uninventoried sources as important contributors (wood combustion, cooking, biogenics, road dust, secondary organic and inorganic aerosol, high emitters)
- Focused emission inventory improvements
- Separated primary emittants from secondary formation products
- 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

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



Receptor modeling has enhanced the concept of using multiple pollutants from multiple sources to assess multiple effects



Chow, J.C.; Watson, J.G. (**2011**). Air quality management of multiple pollutants and multiple effects. *Air Quality and Climate Change Journal,* ***45(3):26-32.***
https://www.researchgate.net/publication/234903062_Air_quality_management_of_multiple_pollutants_and_multiple_effects?ev=prf_pub.

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

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

Lagrangian Source Model

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

- C_{ikl}: CALCULATED AT RECEPTOR
- T_{ijklmn}: CALCULATED BY CHEMICAL MODEL
- D_{kln}: CALCULATED BY MET MODEL
- $F_{ij} Q_{jkmn}$: MEASURED AT SOURCE (INVENTORY)

Chemical Mass Balance (CMB) Model

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

- C_{ikl}: MEASURED AT RECEPTOR
- $T_{ijkl} F_{ij}$: MEASURED AT SOURCE (T=1 OR ESTIMATED BY OTHER METHOD)
- $\Sigma_m \Sigma_n D_{kln} Q_{jkmn}$: S_{ijkl}, SOURCE CONTRIBUTION ESTIMATE

Least squares minimization solutions are often referred to as "the CMB" model, but PMF and UNMIX are also solutions, not separate models

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

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

OWLS-CMB: $\chi^2 = \min \Sigma_i \left[(C_i - \Sigma_j F_{ij} S_j)^2 / \sigma_{Ci}^2) \right]$

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

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

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

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

Positive Matrix Factorization, Paatero (1997), multiple samples

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

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

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

Steps in the applications and validation protocol applies equally well to all of the CMB solutions, but they are rarely followed

1. Determine the applicability of CMB
2. Format input files and perform initial model runs
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

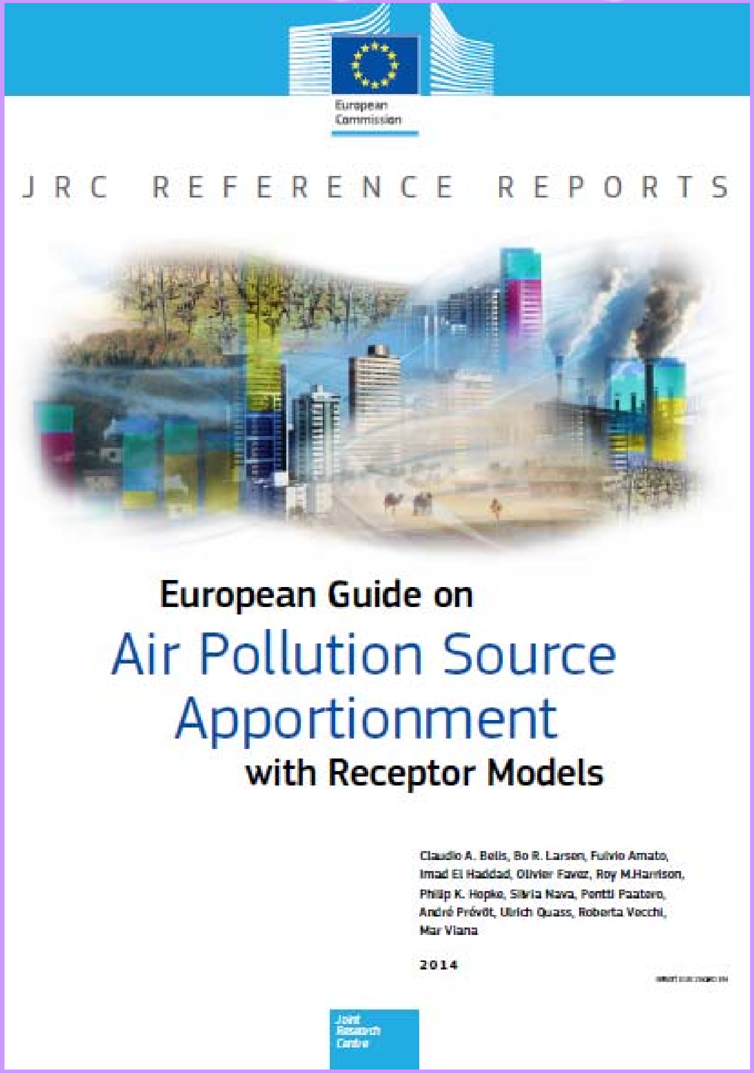
Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze

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

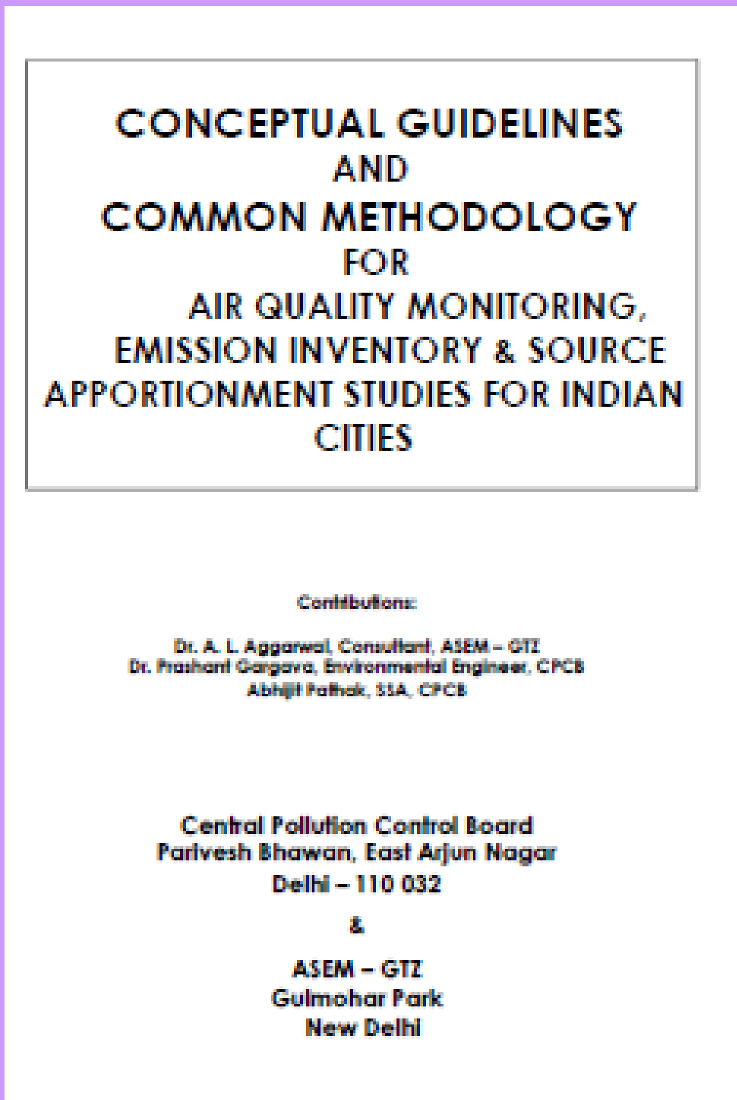
- Examine the problem using different methods
- Use discrepancies between model results to identify and correct weaknesses in models and input data
- Quantify confidence intervals
- Explain and qualify conclusions regarding source contribution estimates

European guidance on receptor modeling is important for introducing more rigor into the source apportionment process



Belis, et al., 2014

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:

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Abhijit Pathak, SSA, CPCB

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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."

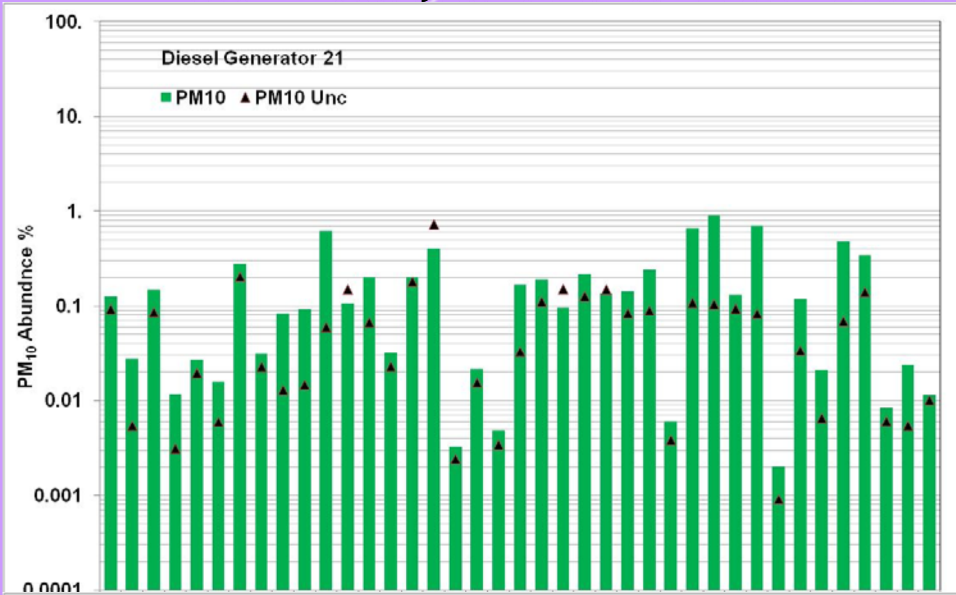
The CPCB study, which Environment Minister Jairam Ramesh has decided not to put in public domain, is likely to be the basis for India's future auto fuel policy. The government has constituted an inter-ministerial group to review the present policy, which expires in 2010, and create one for the new decade.

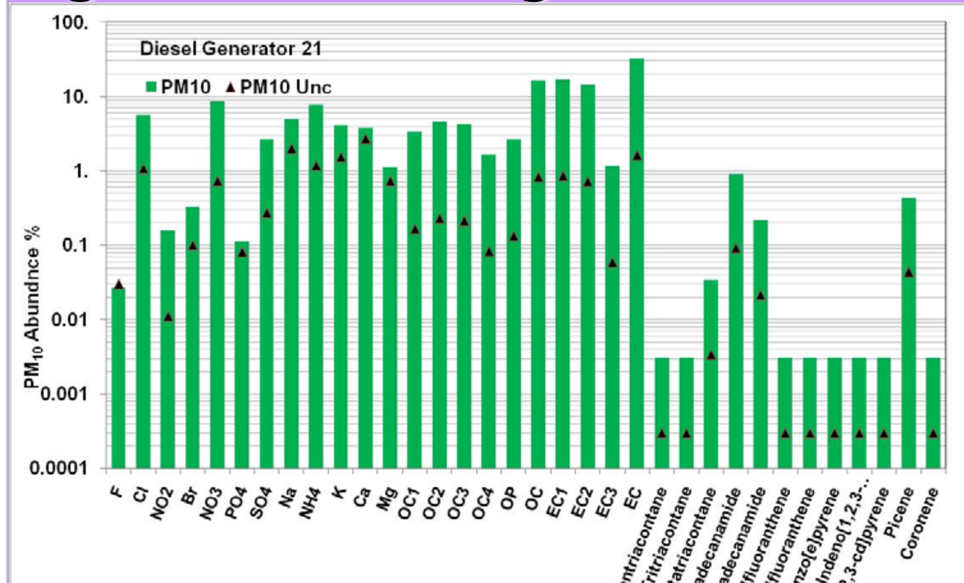
Environment ministry officials said the aim of the new policy would be to reduce the sources of air pollution.

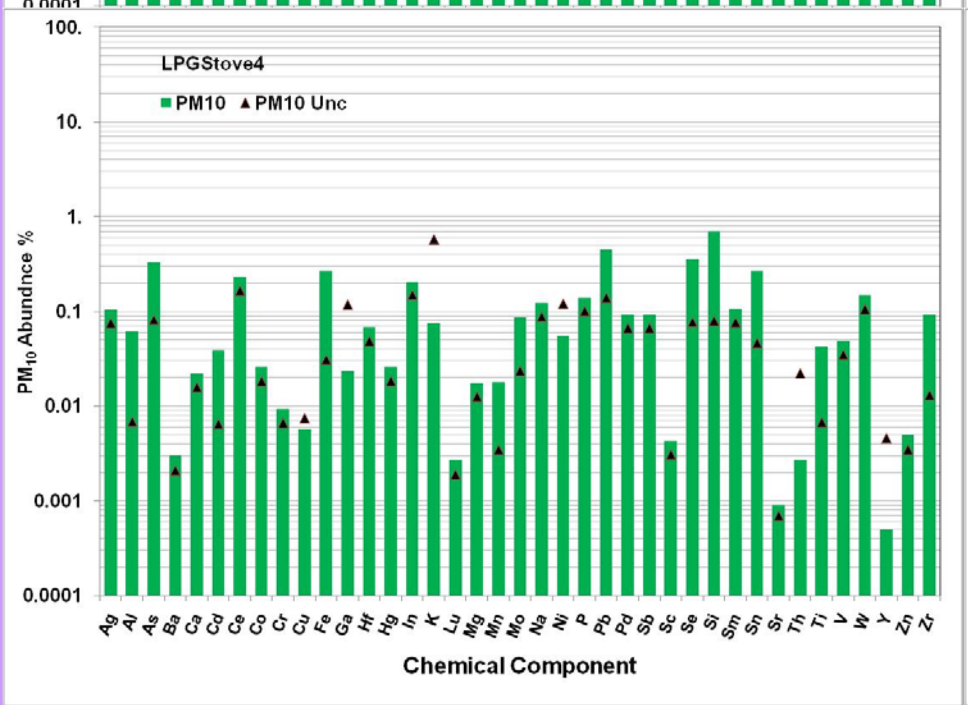
Chowdhury said the government was framing a new policy without consulting people.

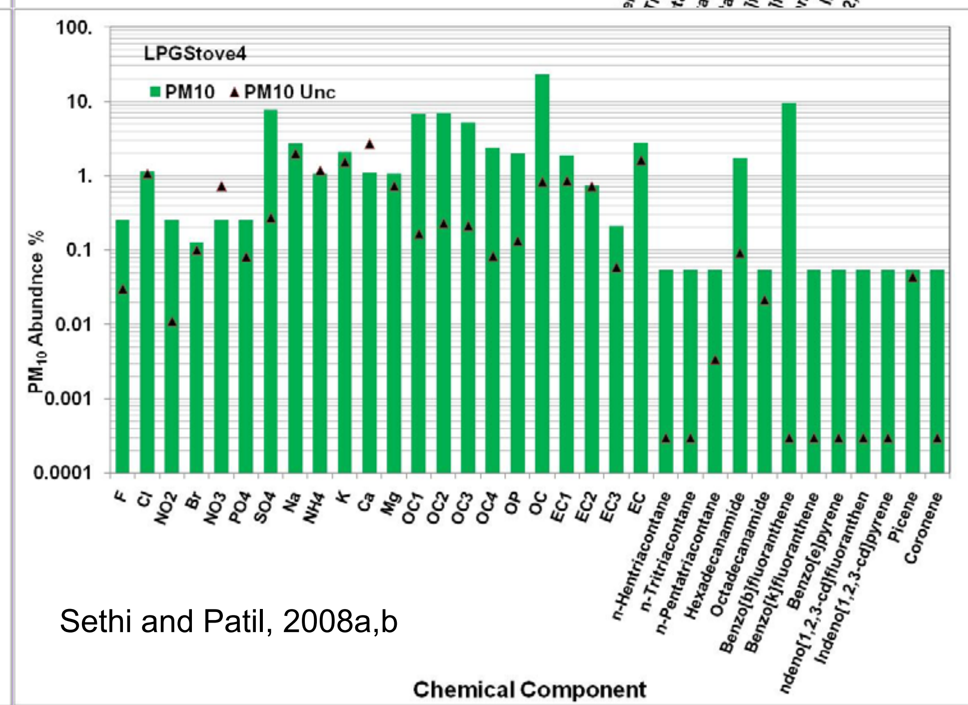
- Weight of evidence would include external data from vehicle and stove emission tests, comparisons with apportionments from different cities, examination of other data such as continuous gas and particle measurements.

Sensitivity tests would have shown that several profiles are collinear, and their source designation must be generalized



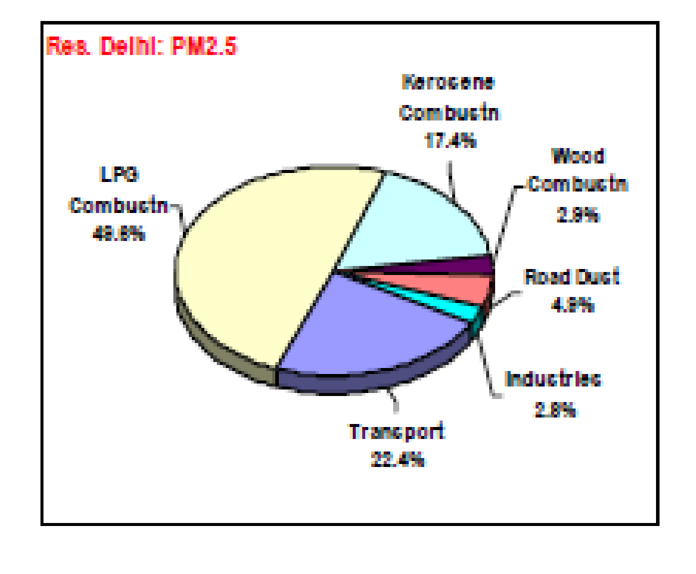


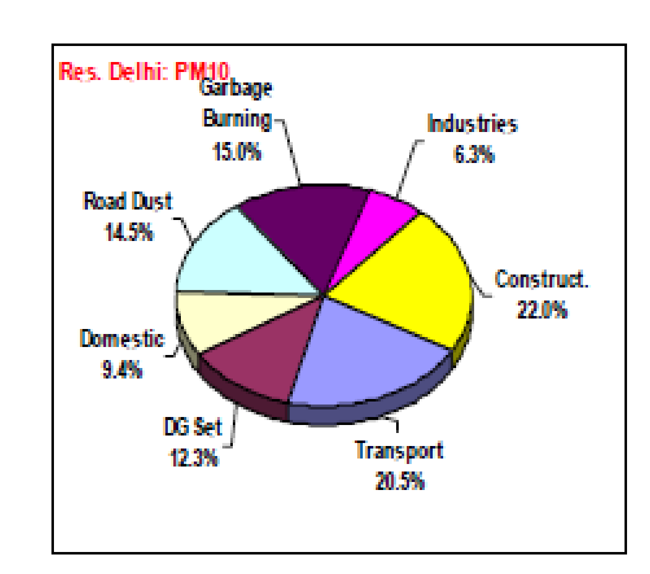


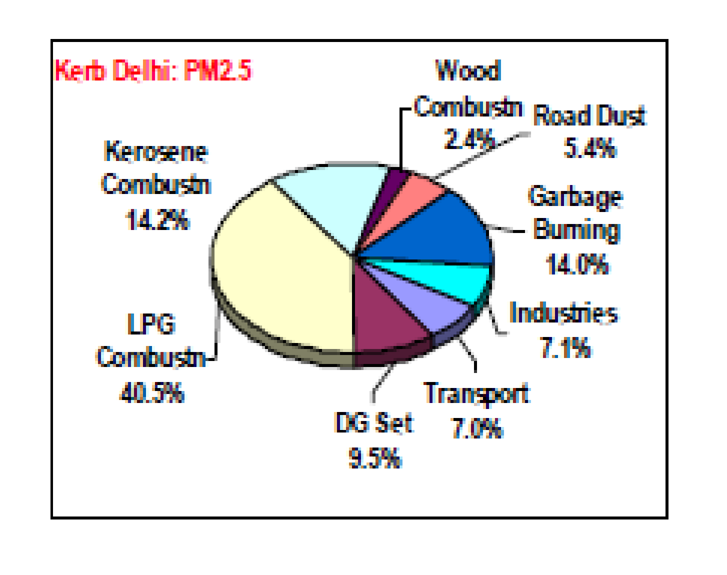


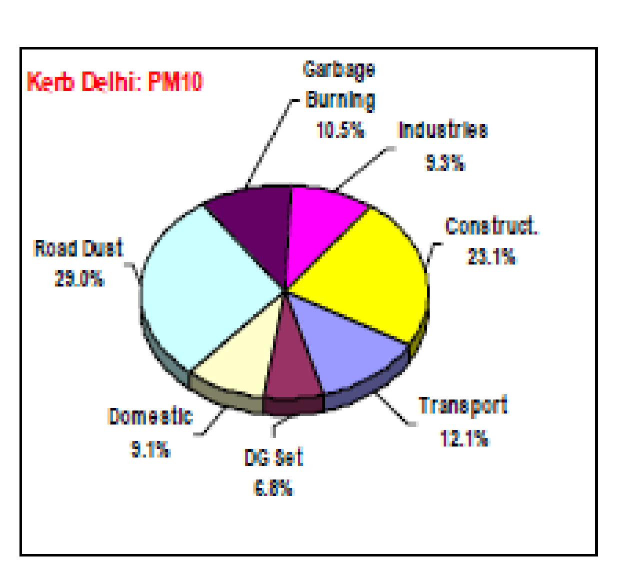
Sethi and Patil, 2008a,b

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



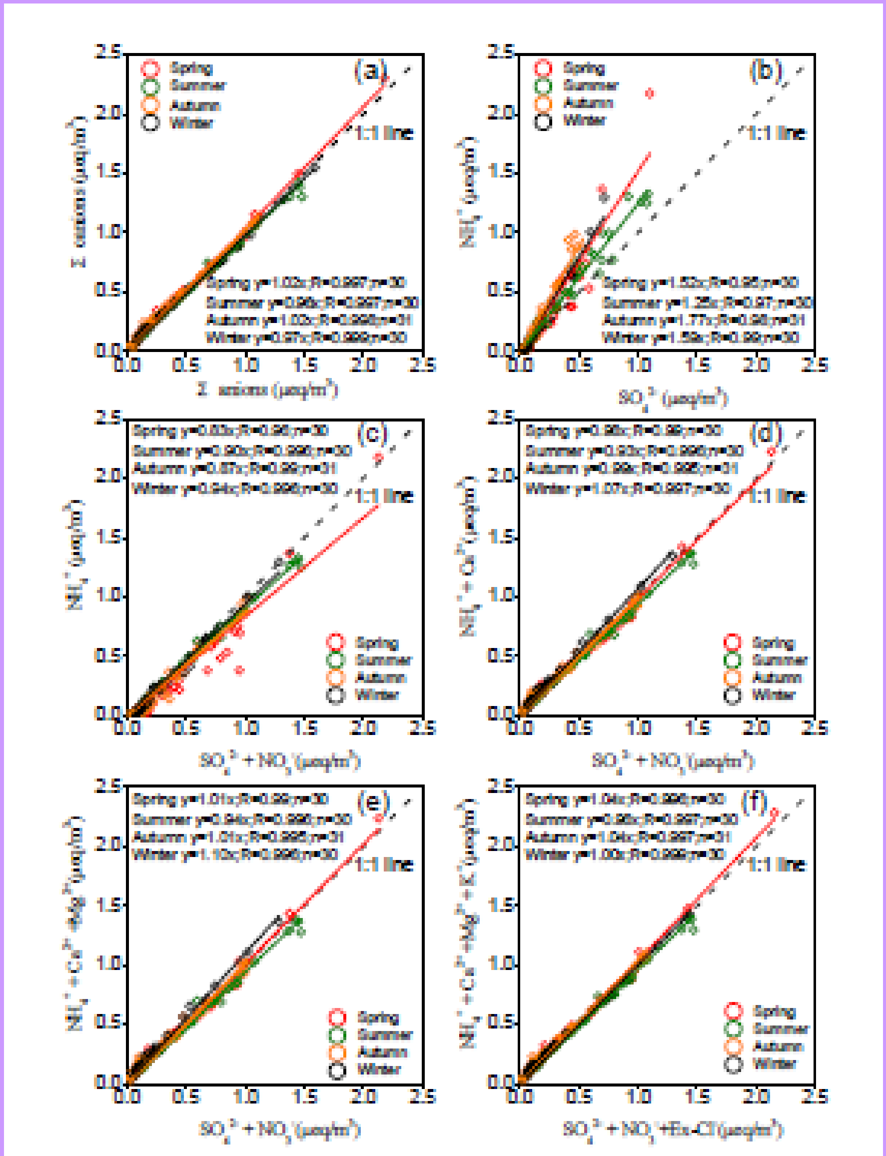






NEERI, 2010

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



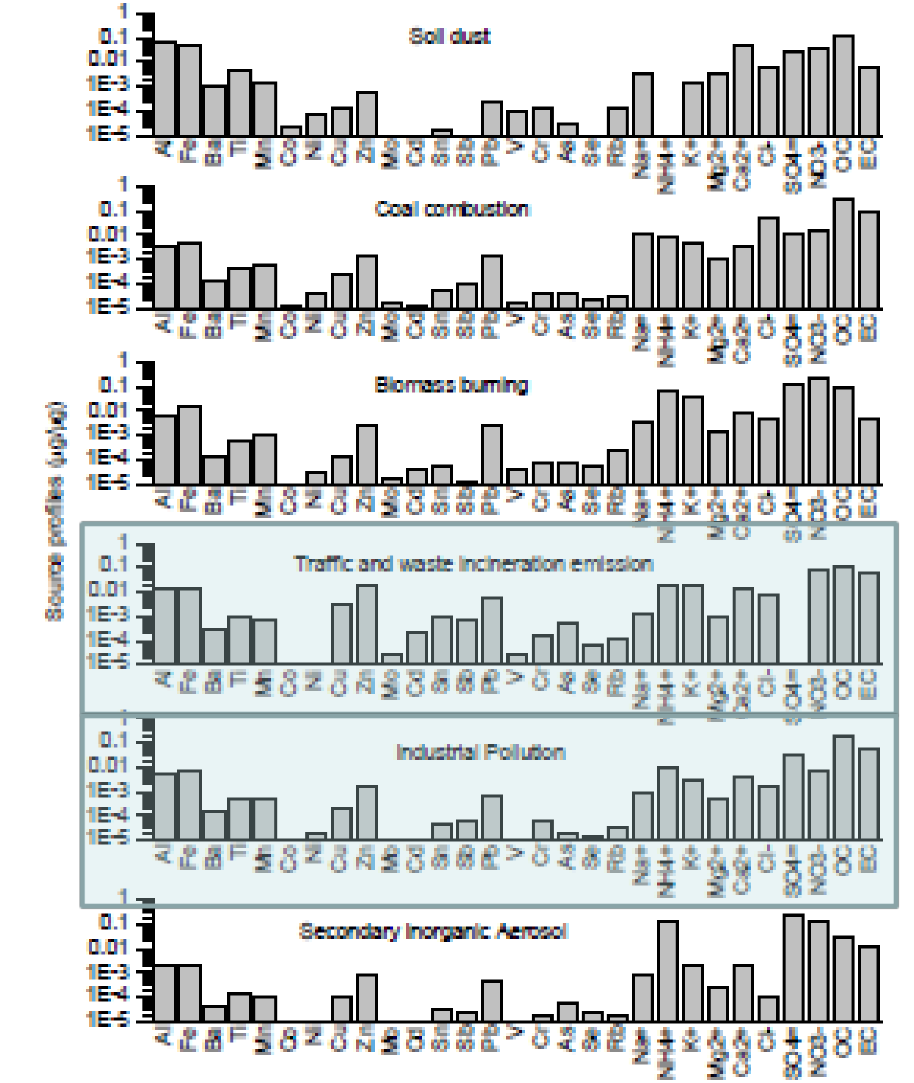
SO_4 was totally neutralized by NH_4, indicating that contributions are more regional than local

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.

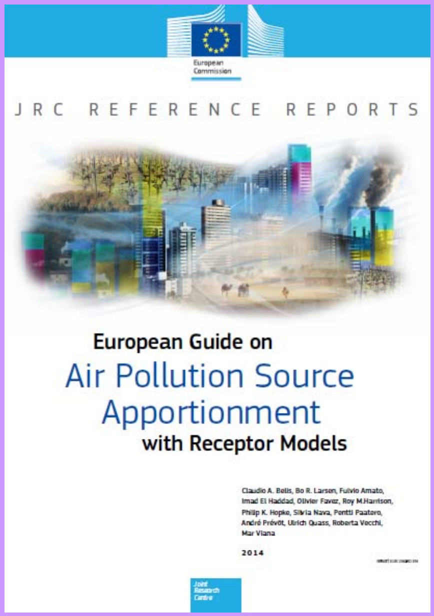
Source	Spring	Summer	Autumn	Winter	Annual
Soil dust	23 %	3 %	18 %	16 %	15 %
Coal combustion	5 %	1 %	7 %	57 %	18 %
Biomass burning	19 %	6 %	17 %	7 %	12 %
Traffic and waste incineration emission	5 %	4 %	4 %	2 %	4 %
Industrial pollution	14 %	32 %	42 %	12 %	25 %
SIA	34 %	54 %	13 %	6 %	26 %



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/.

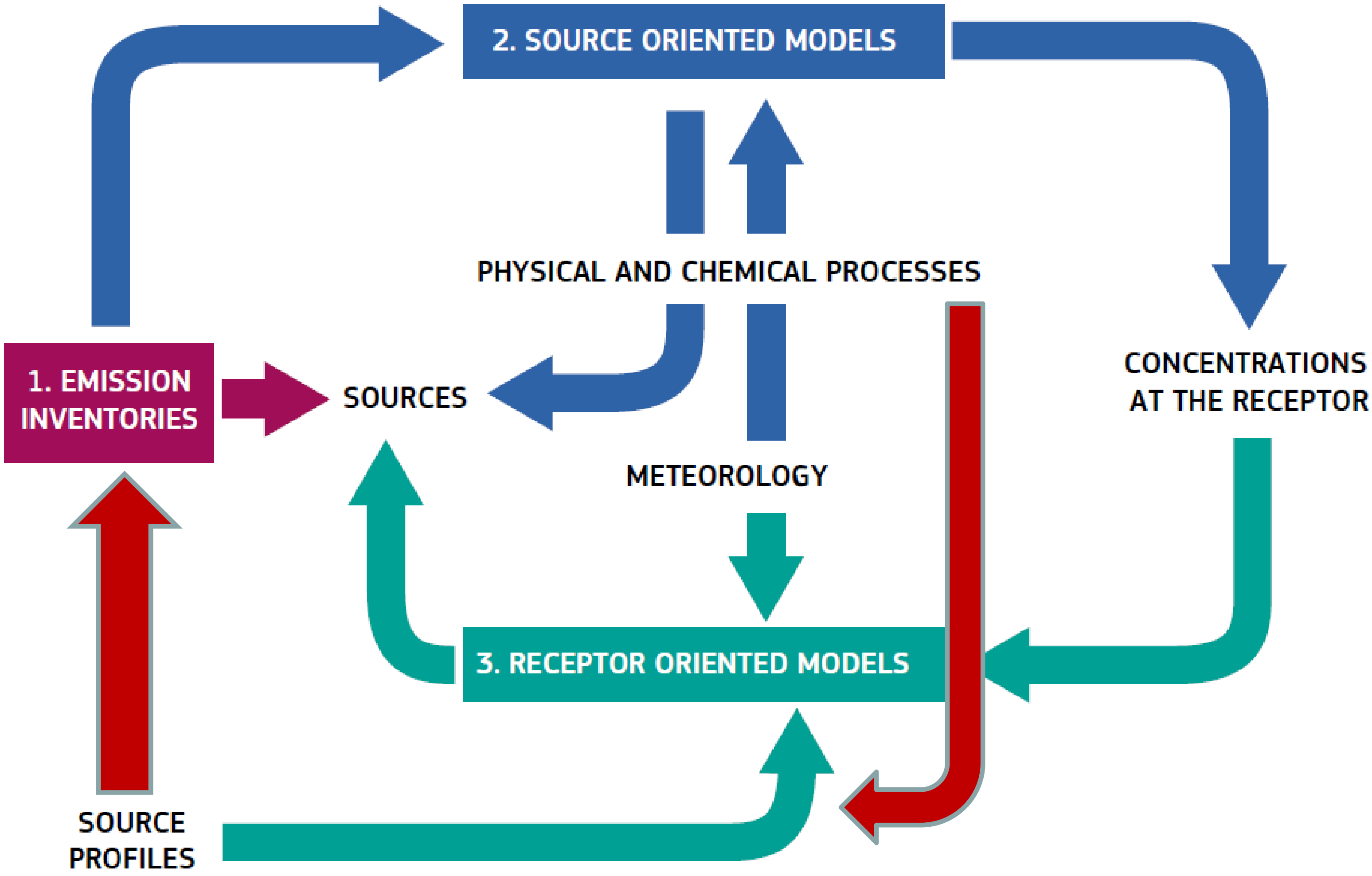
Suggestions for future updates to the European Guide



- Show that most of the methods are different solutions to the CMB equations, and label them as such. (e.g., C^{14}, OC/EC ratios, aethalometer methods are really tracer solutions).
- Be more explicit about importance of network design, i.e. samples from urban-, regional-, and near-source locations.
- Provide more instruction on sensitivity and stability tests
- Elaborate on availability and use of microsensors to accompany filter samplers
- Recommend multiple CMB solutions (e.g., EV, PMF, UNMIX) applied to the same data sets
- Emphasize the need for and practical approaches to obtaining source profiles
- Provide more explicit examples of how receptor models have solved air quality problems

The importance of real-world, multi-pollutant emission rates and source profiles, and their evolution, is under-appreciated

Figure A.1. Schematic representation of the different methods for source identification.



Belis et al., 2014

Table B1 “Common sources of PM in ambient air” is fairly complete for general contributors, but a small number of source sub-types cause excessive levels in Europe and North America

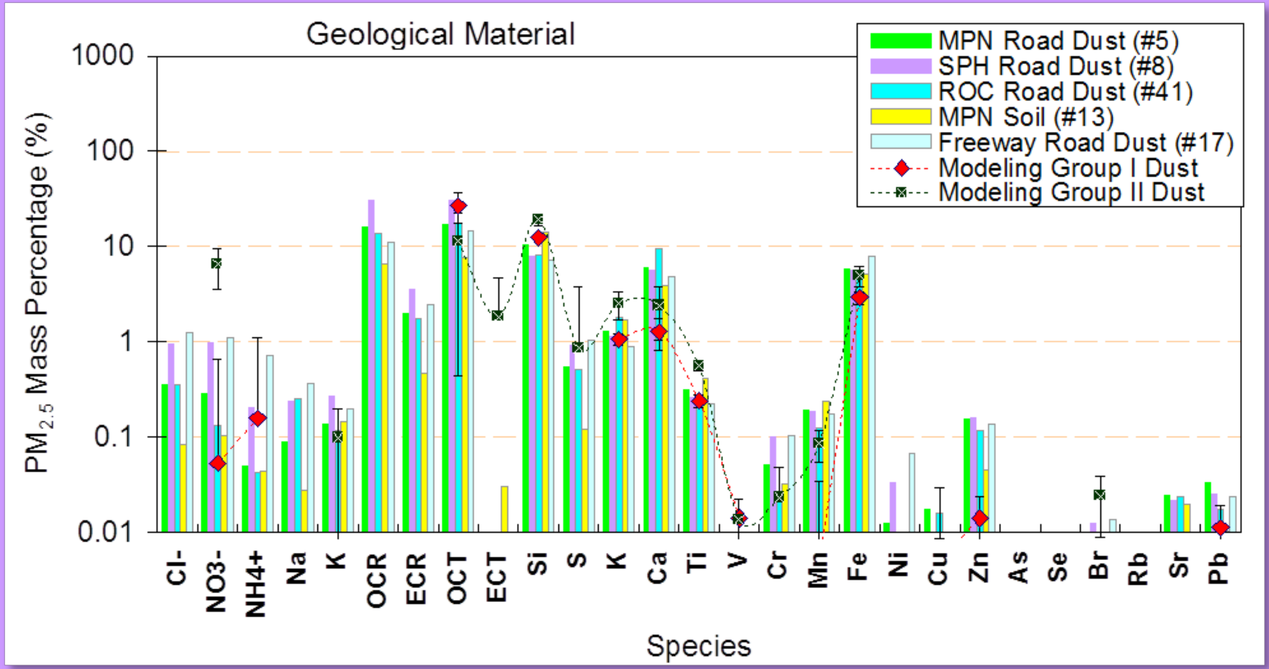
marine salt	industrial emissions
crustal material	secondary ammonium sulphate
road dust	secondary ammonium nitrate
gasoline vehicle exhaust	biomass burning / wood burning
diesel vehicle exhaust	maritime transport
power plants	secondary organic aerosol

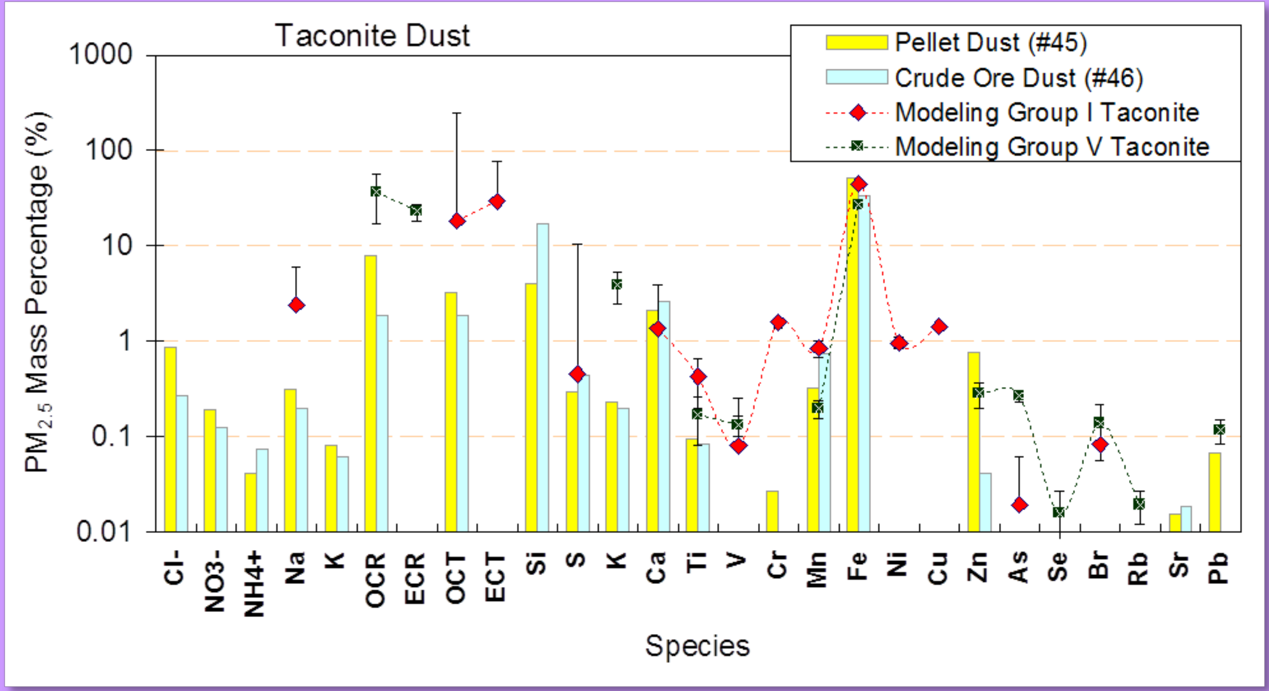
- Primary particles from power plants and industrial sources are usually detectable, but at very low levels
- Wood-burning smoldering and ignition phases often contribute more than the flaming phase
- High emitters of engine exhaust are often important
- Maritime transport means ship engine emissions

Belis et al., 2013

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

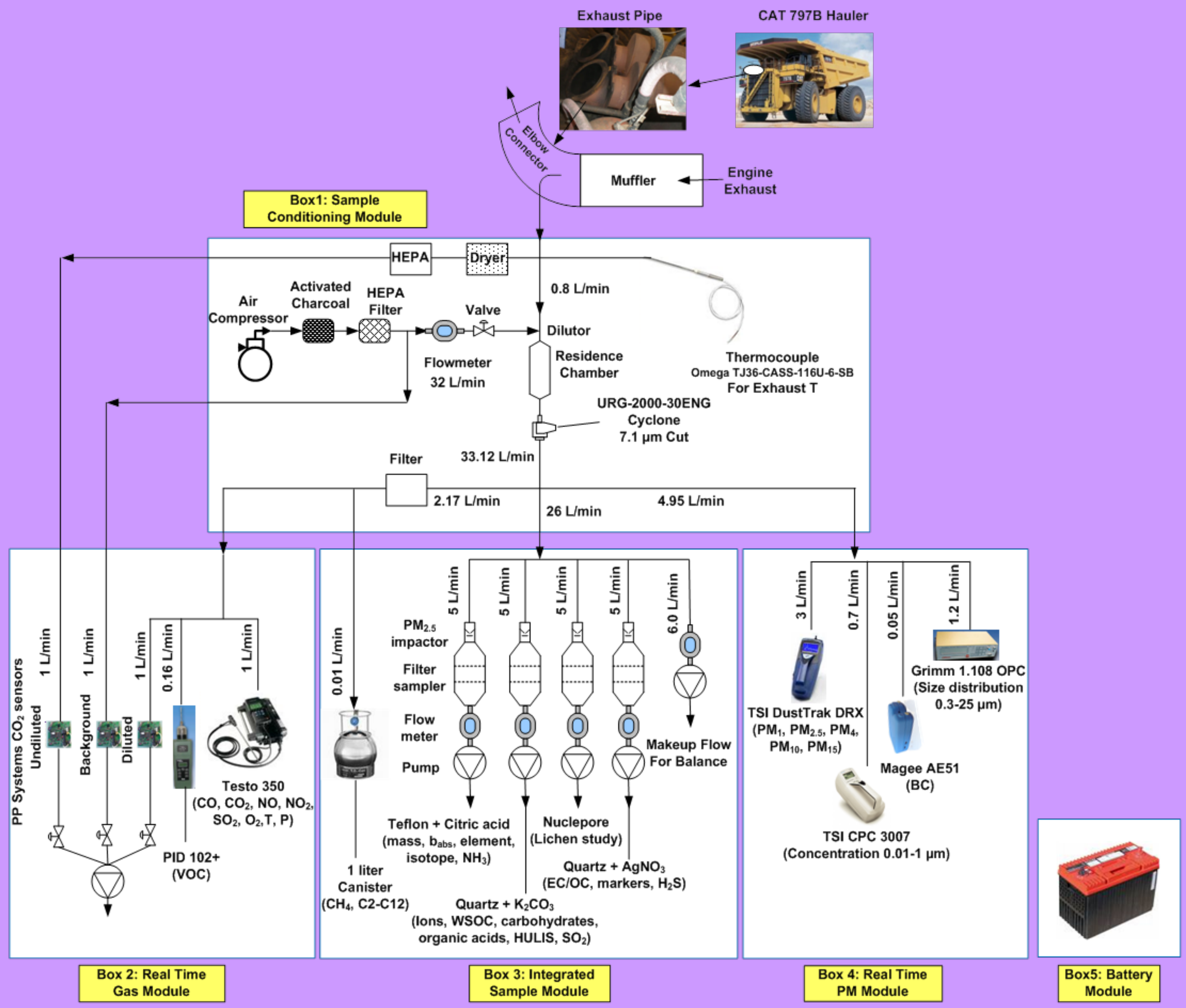
Example from Minnesota



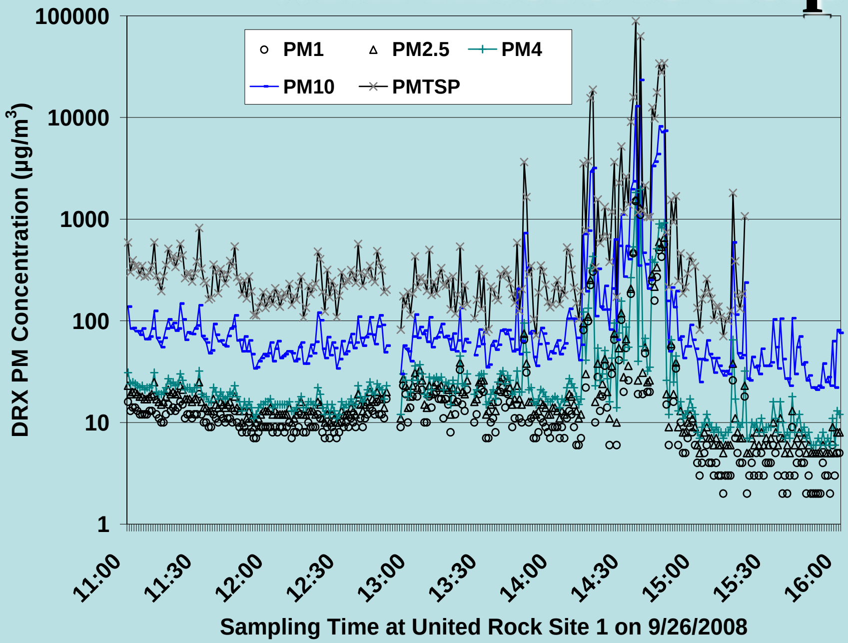


Chen et al. 2010, 2011

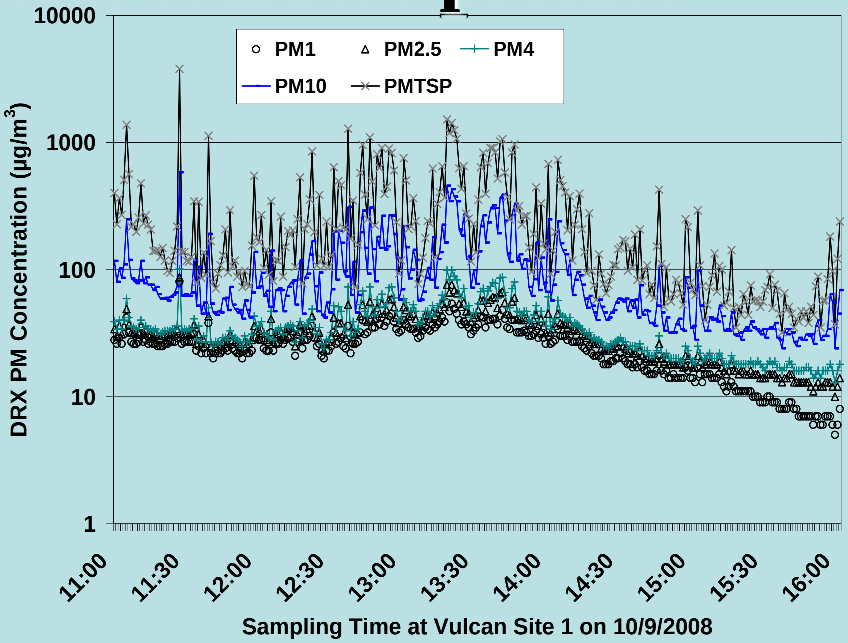
New technologies can be combined into multipollutant systems to obtain source profiles as well as emission rates



Continuous measurements with microsensors can be used with filters to help resolve collinear profiles



Sand / gravel Facility A



Sand / gravel Facility B




* Using TSI DustTrak DRX

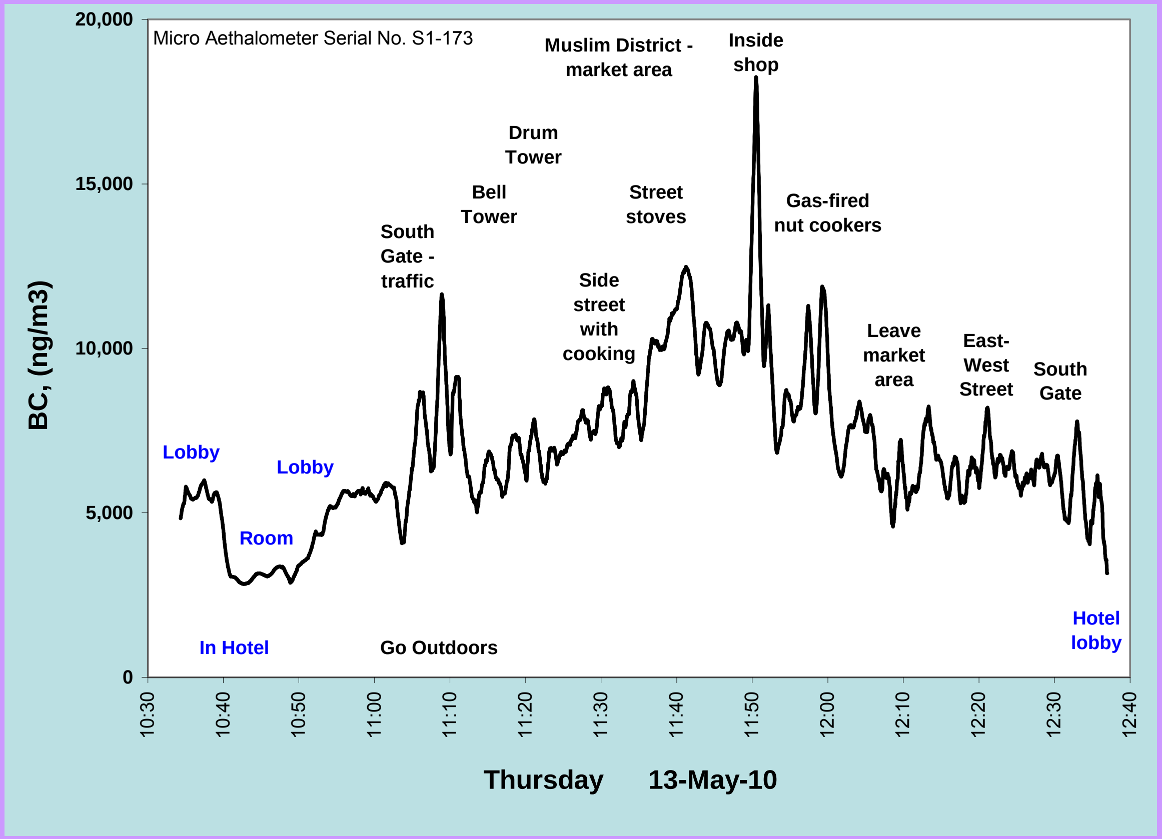




Watson et al., 2011

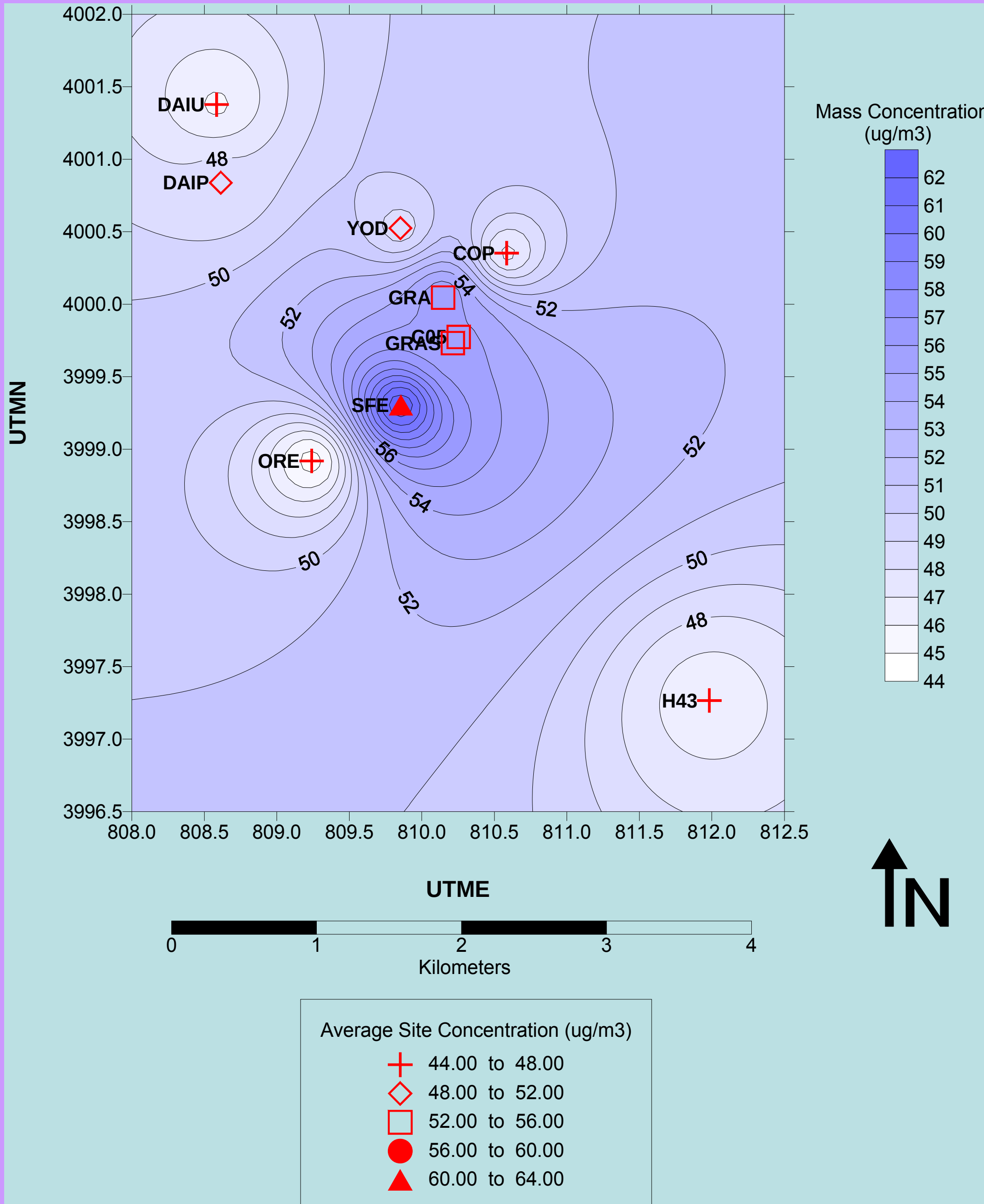
Portable instruments can be used after sampling to determine the zone of influence around a sampling location

(Xian, China)



Hansen and Mocnik, 2010

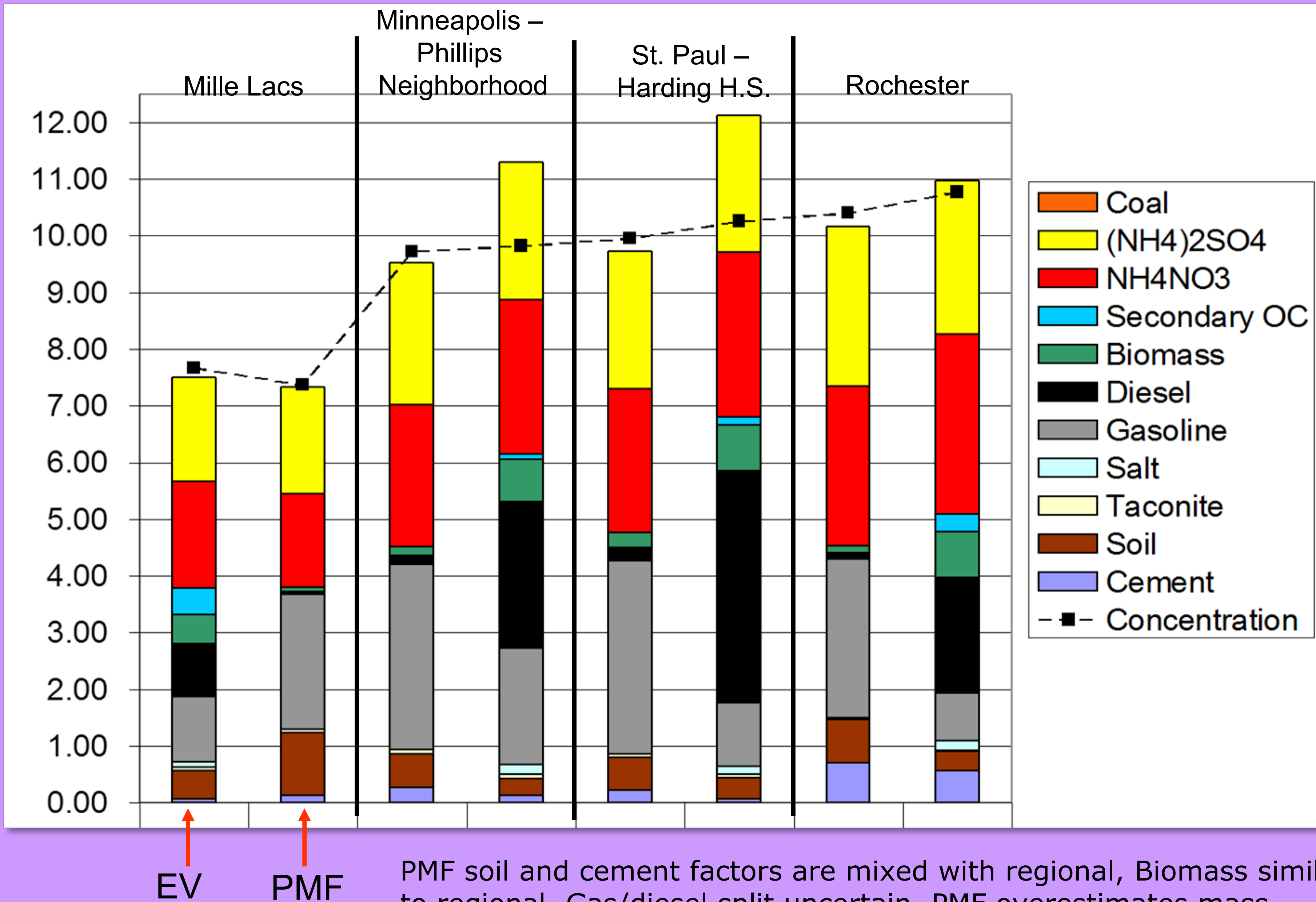
Short-term near-source monitoring can obtain source profiles and better separate PMF factors



Corcoran, central California, USA (10/9/00 – 11/14/00)

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

European examples of receptor models in air quality management need to be added to other examples

- 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)

European examples of receptor models in air quality management need to be added to other examples

- PM_{10} 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 SO_2 scrubbers (Malm et al., 1989)
- Hayden Generating Station SO_2 scrubbers (Watson et al., 1996)
- Mohave Generating Station shutdown (Pitchford et al., 1999)
- Denver Colorado urban visibility standard (Watson et al., 1988)
- Taxi cabs from diesel to natural gas in Hong Kong (Louie et al., 2005a, 2005b)
- California's San Joaquin Valley SIP (Chow et al., 2007)

Conclusions

- Receptor model source apportionment has played a positive role in improving air quality management
- Updated guidance is needed for future applications, especially in developing countries.
- FAIRMODE WG3 seems to be the only authoritative group addressing this issue, and implications for this guidance extend beyond the European situation

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