Black and Organic Carbon Emission Inventories in California

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Objectives

- Describe how and why carbon emission inventories are created
- Examine some of the sources of data
- Identify incompatibilities between source and receptor measurements
- Suggest approaches to improve emission estimates for multiple pollutant/multiple effect air quality management

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Greater detail in:

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Black and Organic Carbon Emission Inventories: Review and Application to California

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ATMOSPHERIC ENVIRONMENT

$PM_{2.5}$ source profiles for black and organic carbon emission inventories

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Science of the Total Environment 409 (2011) 2384–2396

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Science of the Total Environment

Particulate emission factors for mobile fossil fuel and biomass combustion sources

John G. Watson [a,*], Judith C. Chow [a], L.-W. Antony Chen [a], Douglas H. Lowenthal [a],
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Climate Change – Characterization of Black Carbon and Organic Carbon Air Pollution Emissions and Evaluation of Measurement Methods

Phase II: Characterization of Black Carbon and Organic Carbon Source Emissions

DRI Contract Number: 04-307

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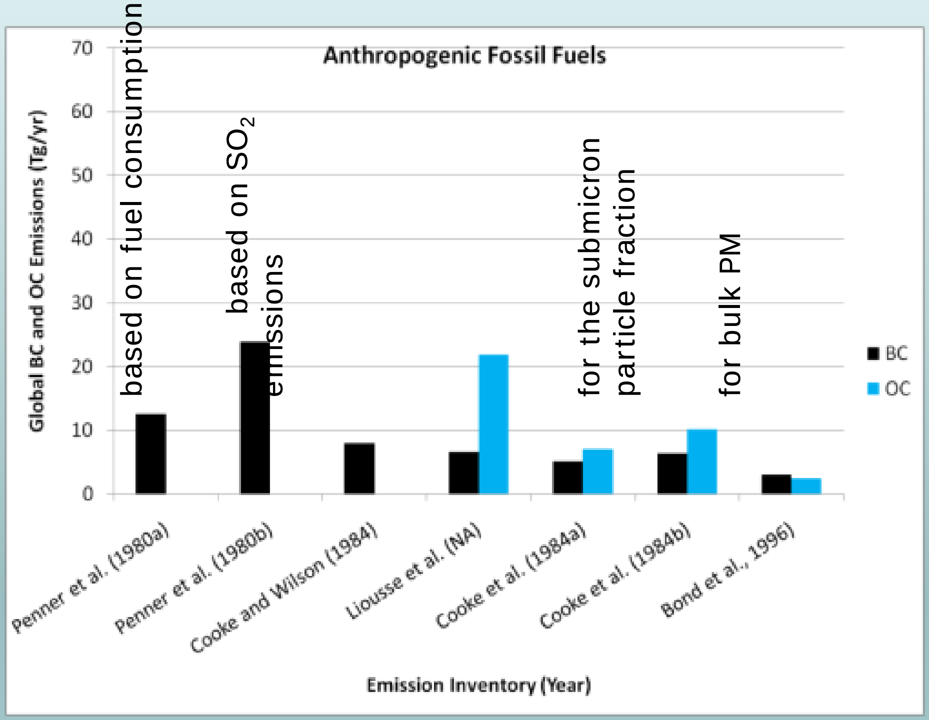
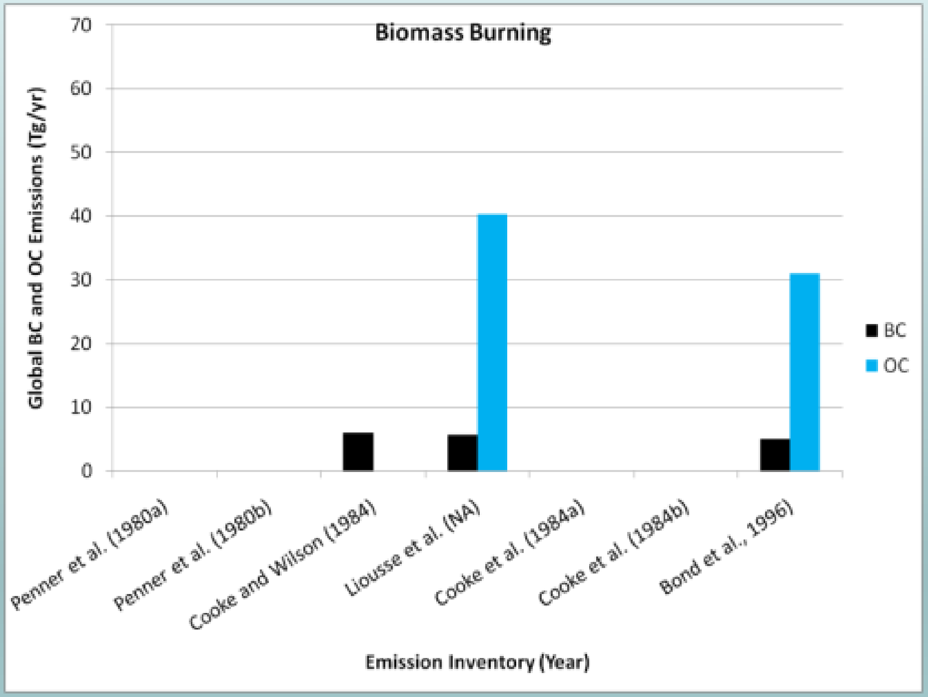
Inefficient fossil fuel combustion and biomass burning are the major global sources of carbon emissions





Sum of fossil fuel and biomass burning
BC: 8 – 14 Tg/yr
OC: 33.4 – 62.2 Tg/yr

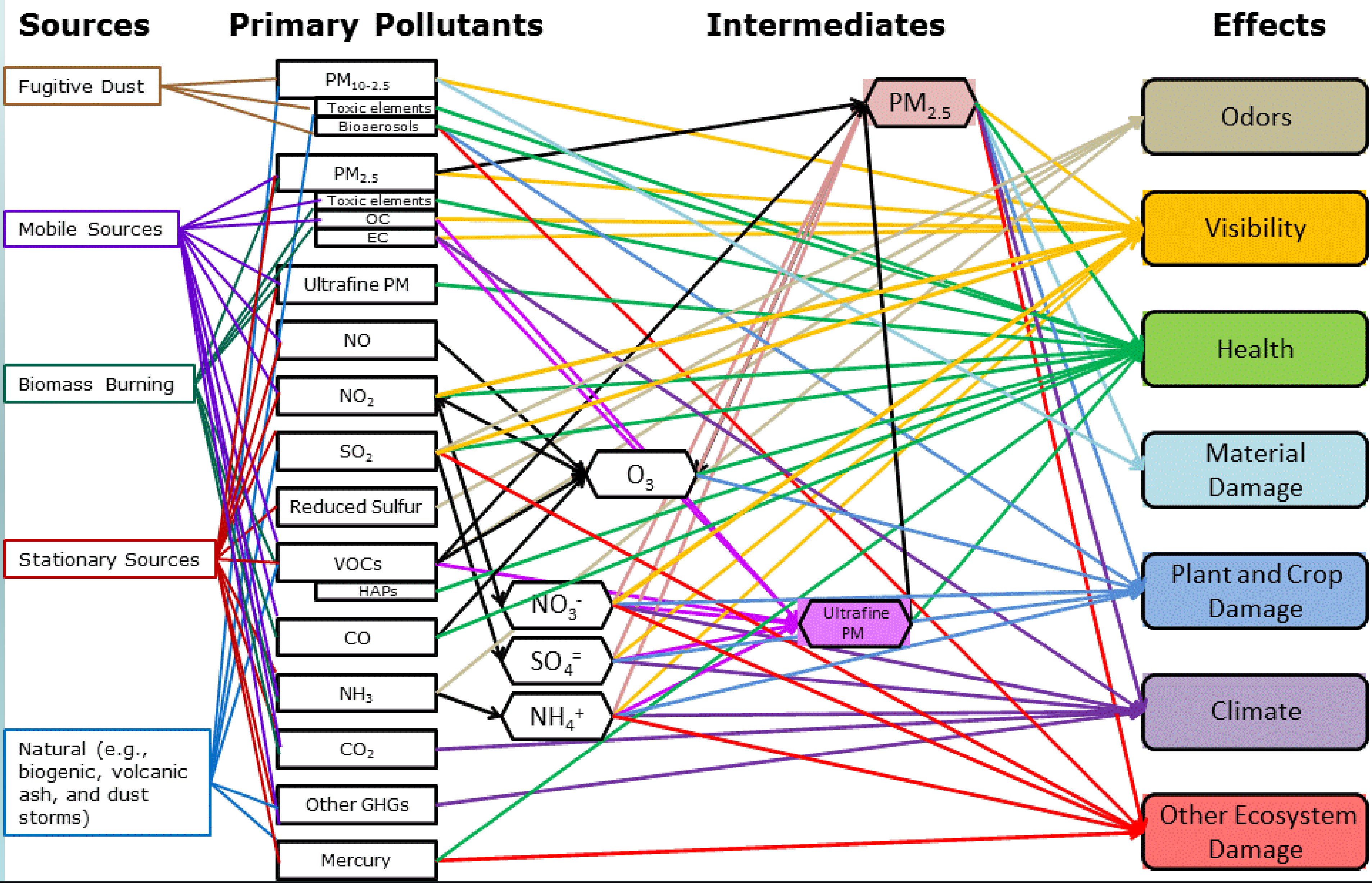
Tg/yr = teragrams/year

While the focus has been on "black carbon", PM comes in many colors that are not black. All of these affect the Earth's radiation balance

One-week duration samples from the Island of Crete (November 2001 to June, 2002) acquired by Helene Cachier.

http://www.wrapair.org/APACE/OCED/Cachier.ppt

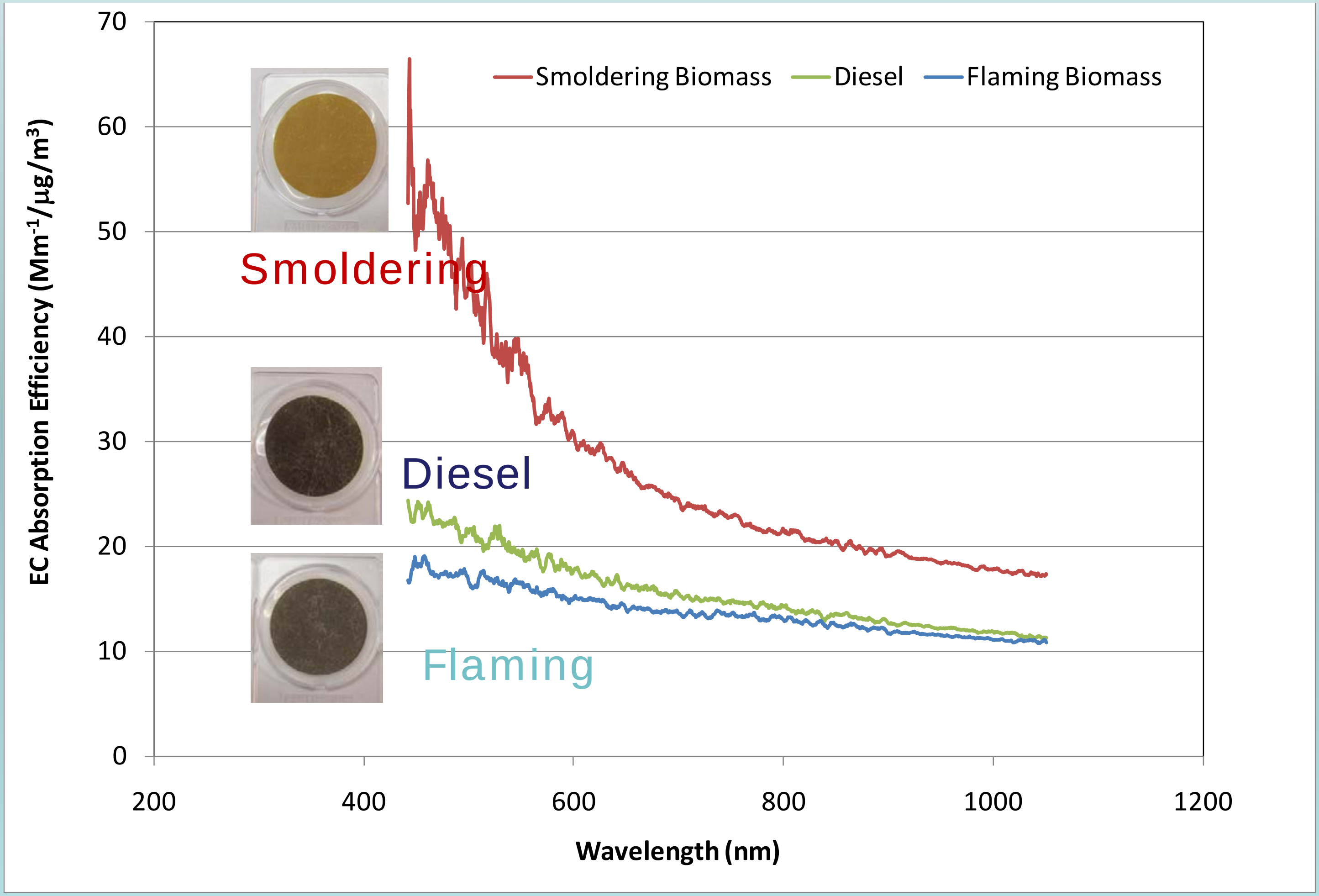
Emissions data are used to determine relationships between multiple sources, pollutants, and effects. Climate is only one of these effects



Chow and Watson,2011

The flaming and smoldering phases of biomass burning show the largest differences and should be separately classified in climate-related inventories

(EC absorption efficiency varies by source and wavelength)



Watson et al., 2011

Emission models combine emission compositions, factors, all of which have large uncertainties

Component i* emissions fluxes =

Σ_{ij} fraction of component i in source j

x emission factor (mass/activity) for source j

x activity of source j

x [particle size fraction]

x [control efficiency]

x [temporal profile]

*i= elemental carbon (EC) or organic carbon (OC)

Watson et al. ,2012

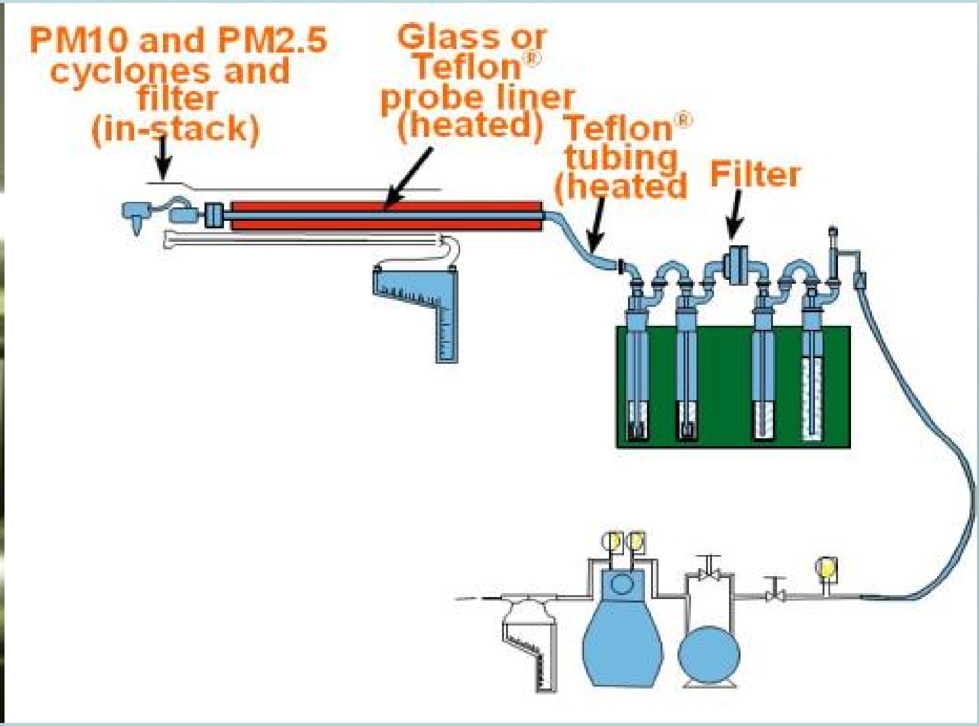
California continues to develop and perfect its emission models

- EMFAC considers on-road engines by technology group and odometer mileage in addition to vehicle model year (http://www.arb.ca.gov/msei/onroad/latest_version.htm)
- First Order Fire Effects Model (FOFEM) that determines the fuel loading characteristics for fuel components by vegetation type. EFs in EES are functions of fuel moisture (i.e., dry, moderate, and wet) and fuel components, including: 1) litter; 2) small wood; 3) large wood; 4) herb and shrub; 5) duff; and 6) canopy fuels. http://www.arb.ca.gov/ei/see/see.htm

PM emissions depend on the test method. Diesel engine certification emission factors differ for the same operating conditions on fuels depending on whether they are classified as mobile or stationary sources

Mobile emissions are tested on dynamometers with dilution and cooling prior to measurement

Stationary source testing samples hot exhaust

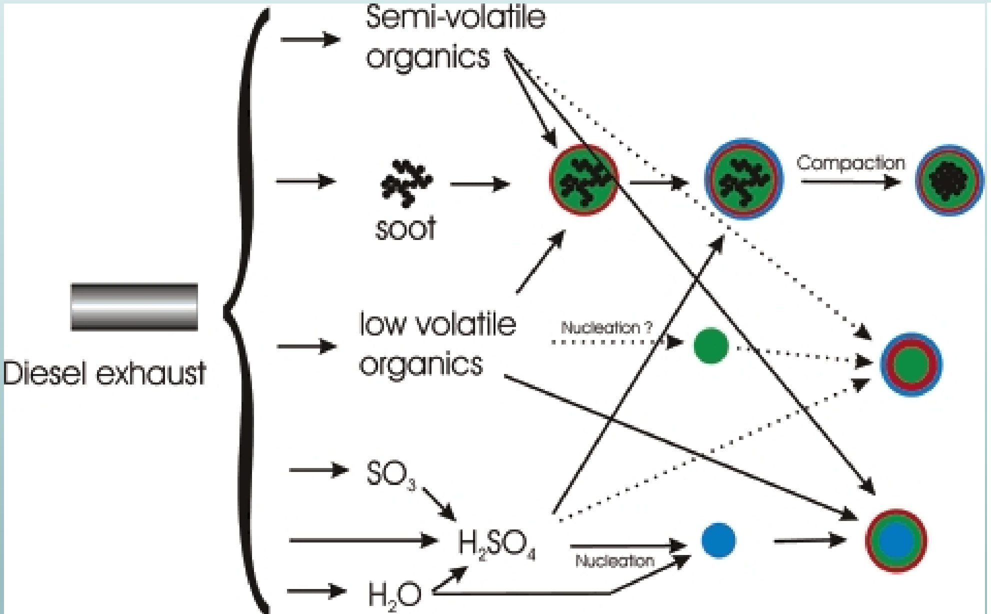


Put generator on wheels and move it and it is certified by dilution sampling. Install the generator permanently and it is certified by hot stack sampling and yields different emissions

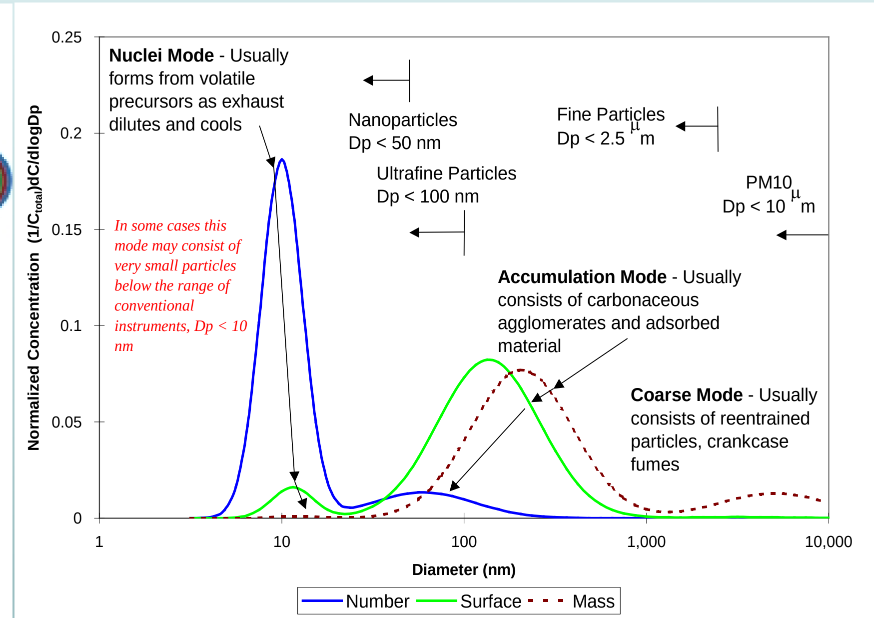
Watson et al., 2012

Cooling, dilution and aging leads to particle nucleation and growth

Particle evolution



Particle size distribution



Factors Affecting Diesel Emissions:

- Engine types and power
- Engine operating conditions (e.g., idle, accelerate, and decelerate)
- Fuel formulations (e.g., sulfur or aromatic content)
- Dilution and aging
- Meteorology (e.g., sunlight, temperature, and relative humidity)
- Interactions with ground-level environment

Kittelson (1998)
Schneider et al. (2005)

PM organic carbon and sulfates form at lower temperatures in diluted ship stack emissions

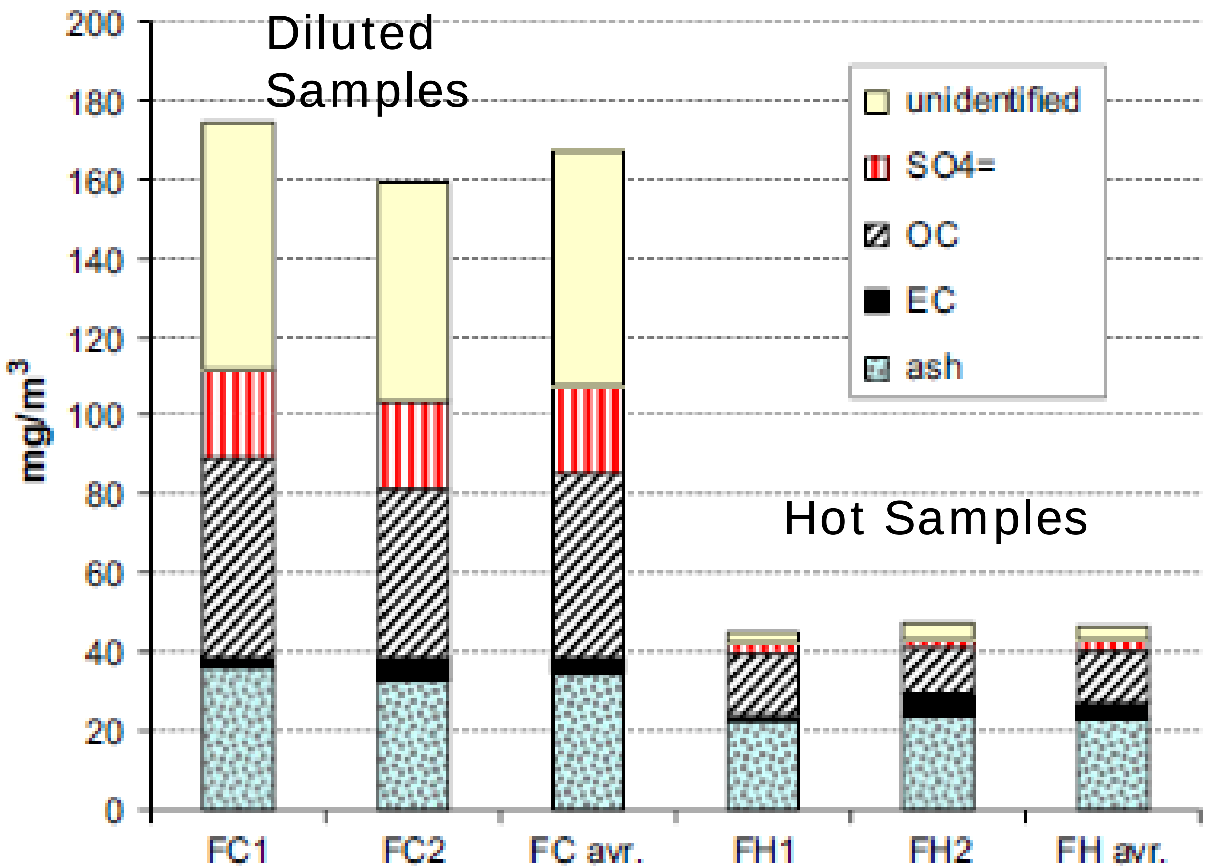
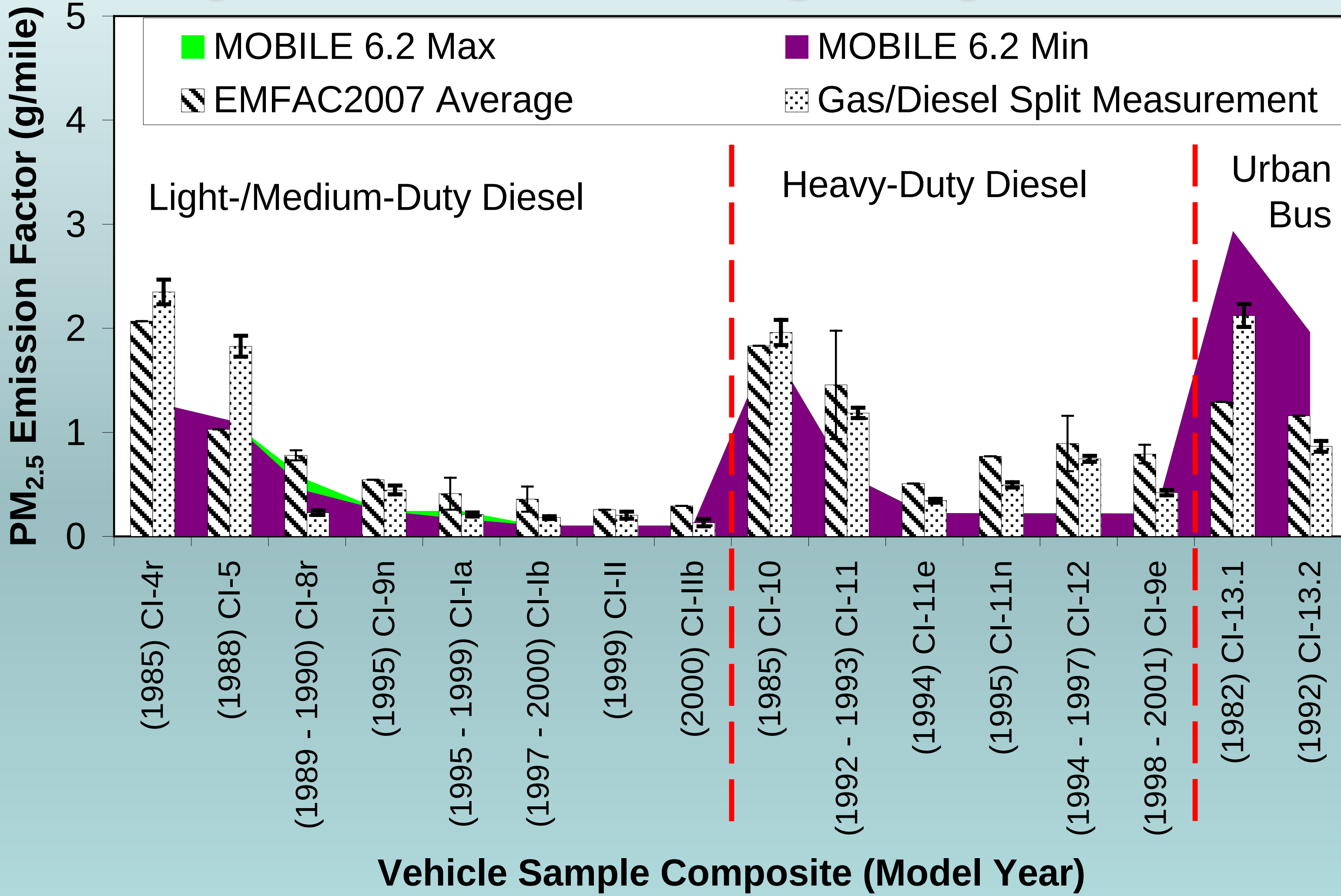


Fig. 3. Composition of PM (as mg m^{-3} exhaust gas) collected on filters in the diluted (FC) and hot (FH) exhaust gas. FC1, FC2, FCavr. and FH1, FH2, FHavr. are individual filter samples and their average values collected in the diluted and hot exhaust, respectively.

Moldanova et al., 2009

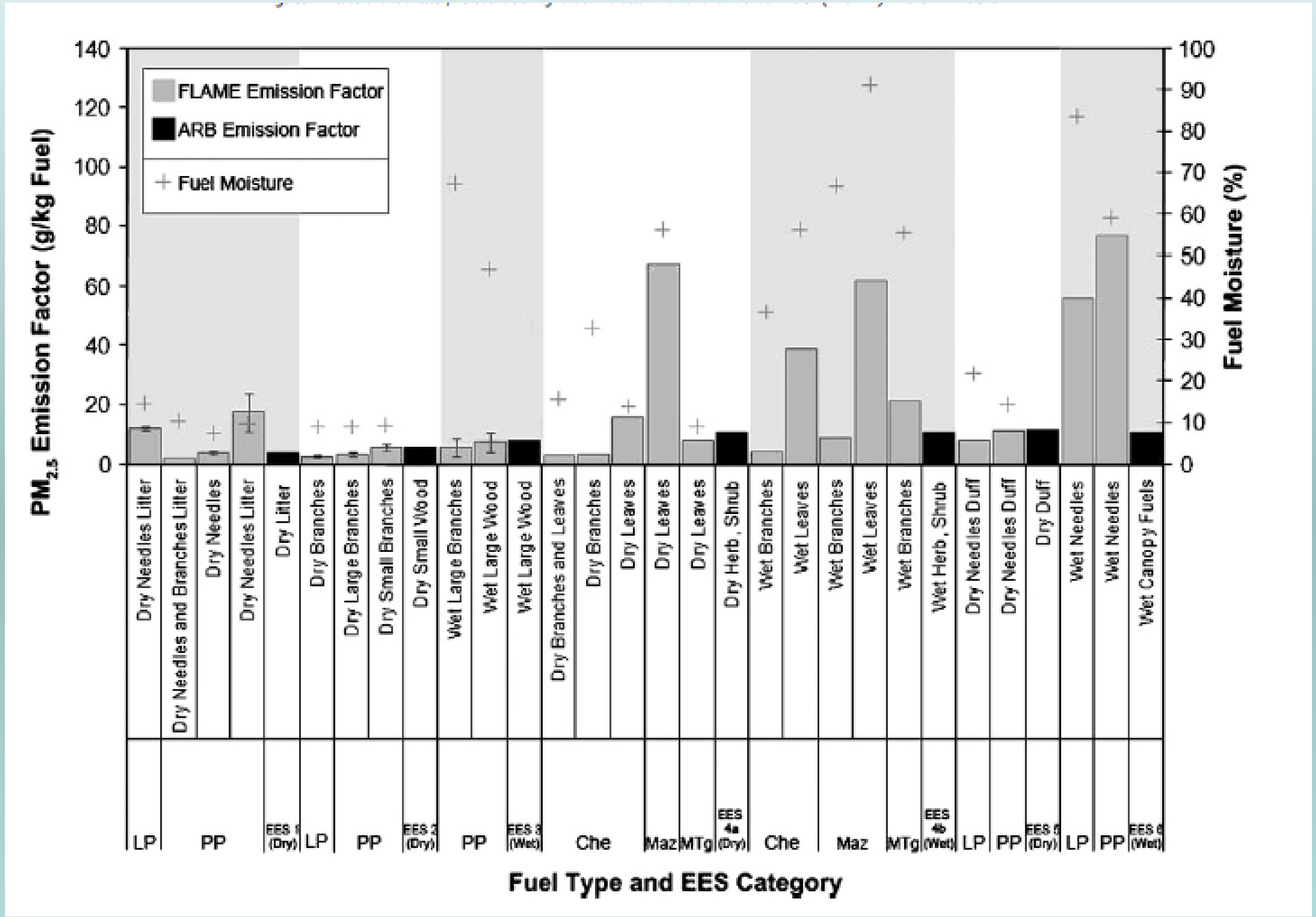
PM emission factor models for on-road engines in developed countries are improving, but these are becoming less important emitters with engine improvements



Hot City-Suburban route (HCS) driving cycle during the Gas/Diesel Split Study with MOBILE 6.2 and EMFAC 2007 model estimates for the Federal Test Procedure (FTP) cycle.

Fujita et al., 2007, 2012, Watson et al., 2011

Fuel moisture is an important variable in biomass burning that is not easily incorporated into emissions models

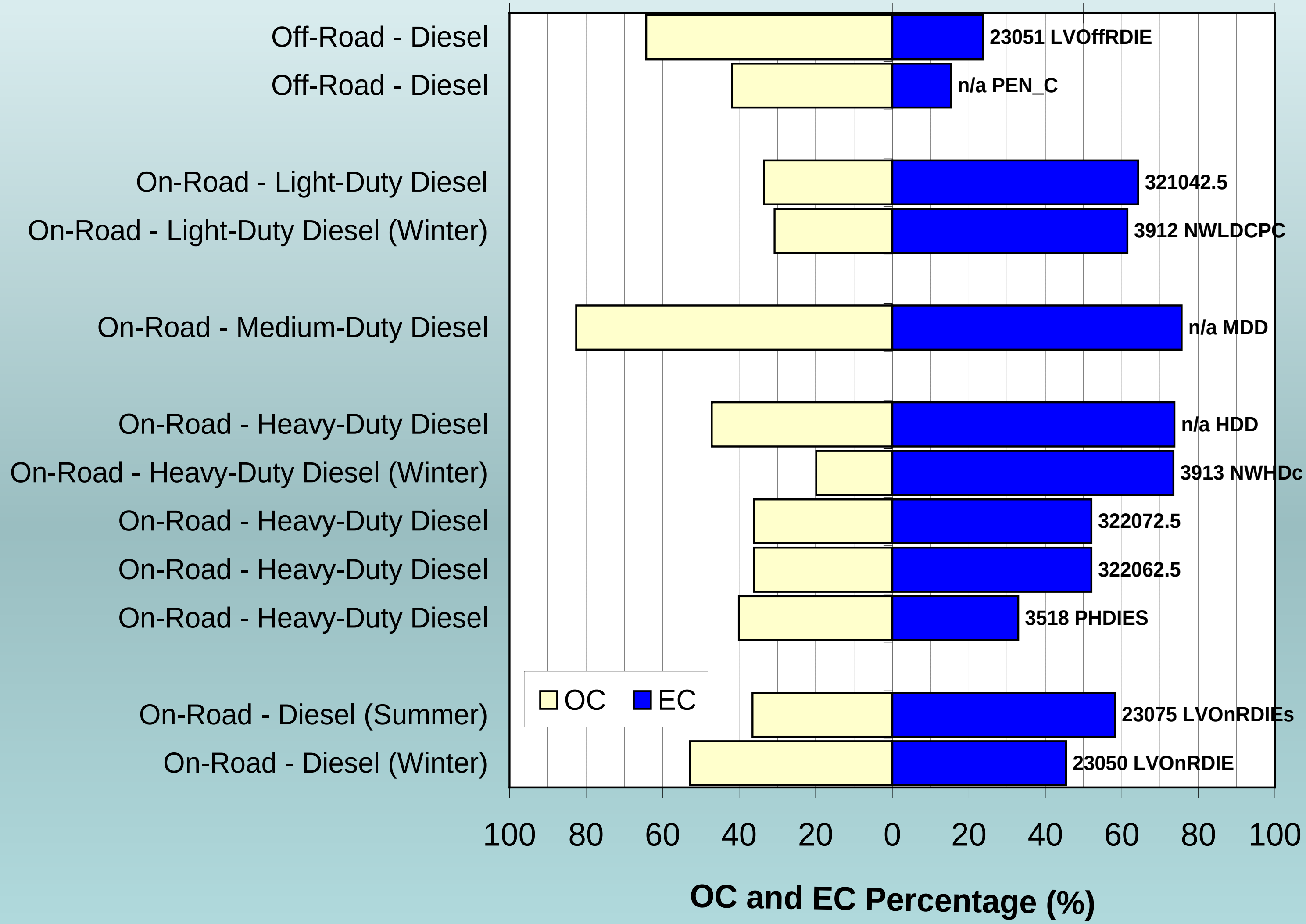


Watson et al., 2011

Chemical source profiles are determined only for special studies and are compiled in data bases

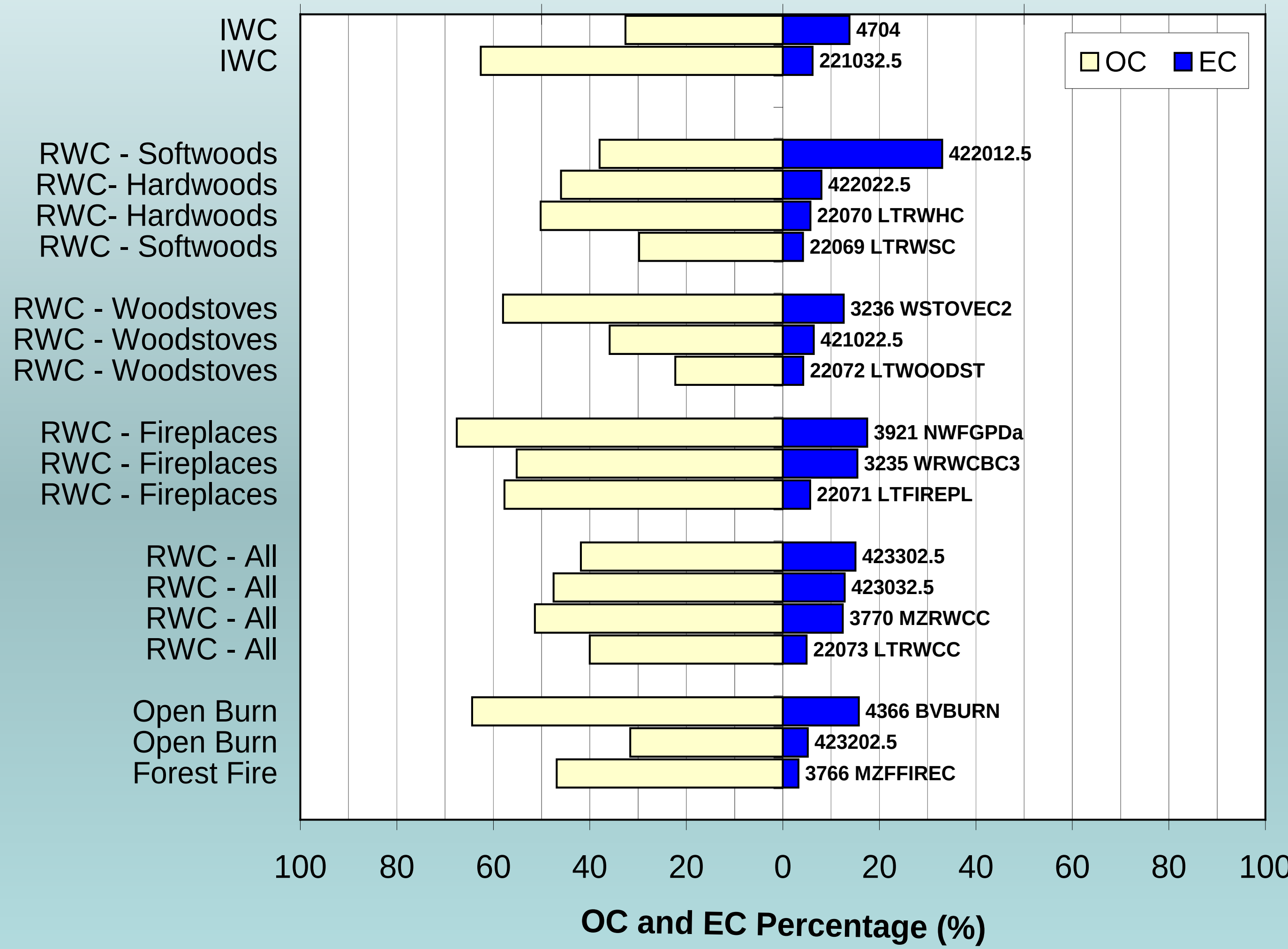
- ARB speciation profiles http://arb.ca.gov/ei/speciate/speciate.htm
- EPA SPECIATE profile data base http://www.epa.gov/ttn/chief/software/speciate/index.html
- Both are pull (periodically adding profiles) rather than push (accepting submissions and descriptions from researchers) data sets and are often out of date

$PM_{2.5}$ OC and EC abundances are highest in diesel exhaust from older engines, and they are highly variable



Chow et al., 2011

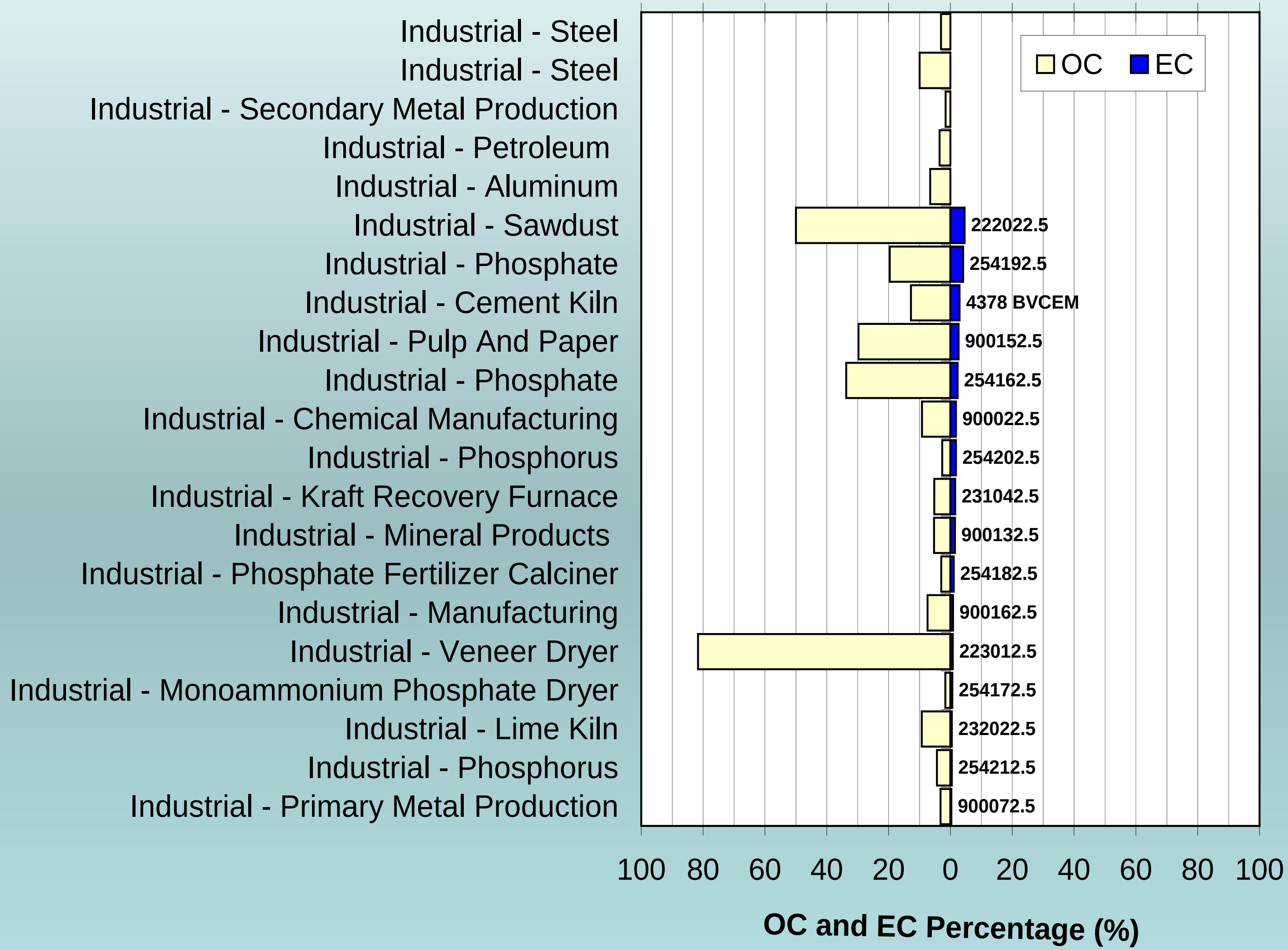
$PM_{2.5}$ OC and EC abundances are even more variable for biomass burning



IWC: Industrial Wood Combustion; RWC: Residential Wood Combustion

Chow et al., 2011

OC and EC abundances are generally low in industrial stack emissions from developed countries



Chow et al., 2011

BC abundances in source samples also depend on the measurement method and are not compatible with ambient measurements

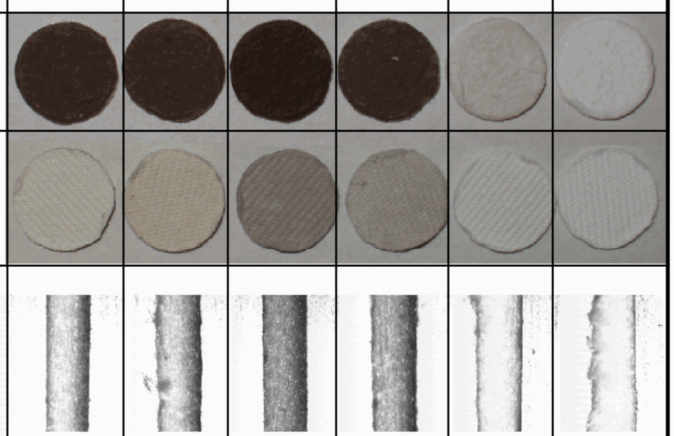
Instruments	Operating principle	Observables	Avg time
Dual wavelength Aethalometer (370, 880 nm)	Filter-based light attenuation	Light absorption (Mm^{-1}) or BC ($\mu g/m^3$)	5 min
Seven color Aethalometer (370, 450, 571, 615, 660, 880, and 950 nm)	Filter-based light attenuation	Light absorption (Mm^{-1}) or BC ($\mu g/m^3$)	5 min
PSAP (467,530,and 660 nm)	Filter-based light attenuation	Light absorption (Mm^{-1}) or BC ($\mu g/m^3$)	5 min
MAAP (670 nm)	Filter-based light attenuation with compensating light scattering effects	Light absorption (Mm^{-1}) or BC ($\mu g/m^3$)	5 min
DMT Photoacoustic (405, 532, and 781 nm)	Light absorption of particles in air based on heating and cooling that creates a sound wave	Light absorption (Mm^{-1}) or BC ($\mu g/m^3$)	5 min
Sunset carbon analyzer (660 nm)	Thermal/optical transmittance (TOT; NISOH 5040 protocol)	EC and OC ($\mu g/m^3$) and optical BC ($\mu g/m^3$)	1-24 hour
R&P 5400 carbon analyzer	Thermal OC/EC at 275 °C and 750 °C	EC and OC ($\mu g/m^3$)	1 hour
PAS 2000 PAH monitor	Photoionization	Particle bound PAH (fA)	5 min
DRI carbon analyzer	Thermal/optical reflectance (TOR; IMPROVE_A protocol)	EC and OC ($\mu g/m^3$)	1-24 hr

Chow et al.2009

OC turns into EC during thermal analyses

- OC in the aerosol deposit on the filter surface chars during thermal analysis
- Adsorbed organic vapors also char filter during analysis, causing differences between reflectance and transmittance measurements

Sample 12/25/02 in IMPROVE Protocol

Temperature (°C) and Combustion Atmosphere	50 (He)	250 (He)	549 (He)	550* (He/O_2)	550** (He/O_2)	800 (He/O_2)
Reflectance (R)	588	582	516	575	1314	1561
Transmittance (T)	6	4	0	0	268	538
Front						
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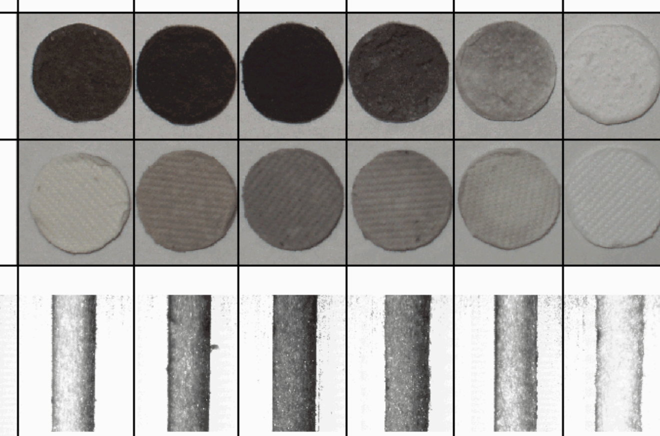
* Beginning of EC1 stage.

** End of EC1 stage.

Sample 12/25/02 in STN Protocol

Temperature (°C) and Combustion Atmosphere	50 (He)	609 (He)	897 (He)	644 (He/O_2)	677 (He/O_2)	920 (He/O_2)
Reflectance (R)	587	507	365	573	897	1564
Transmittance (T)	5	0	0	0	4	534
Front						
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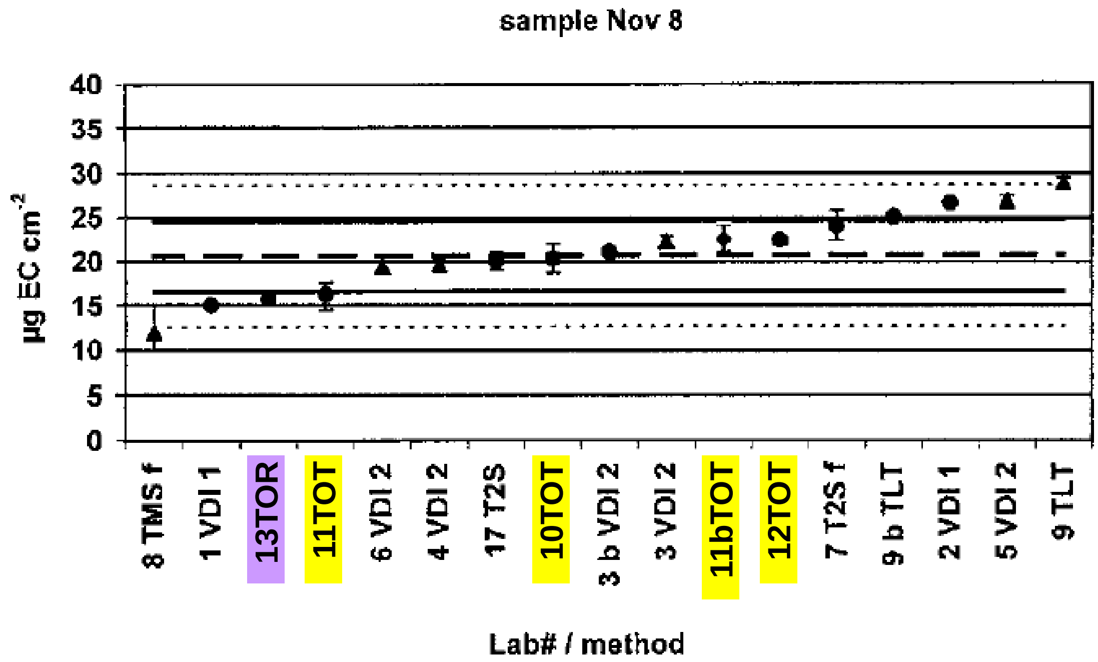
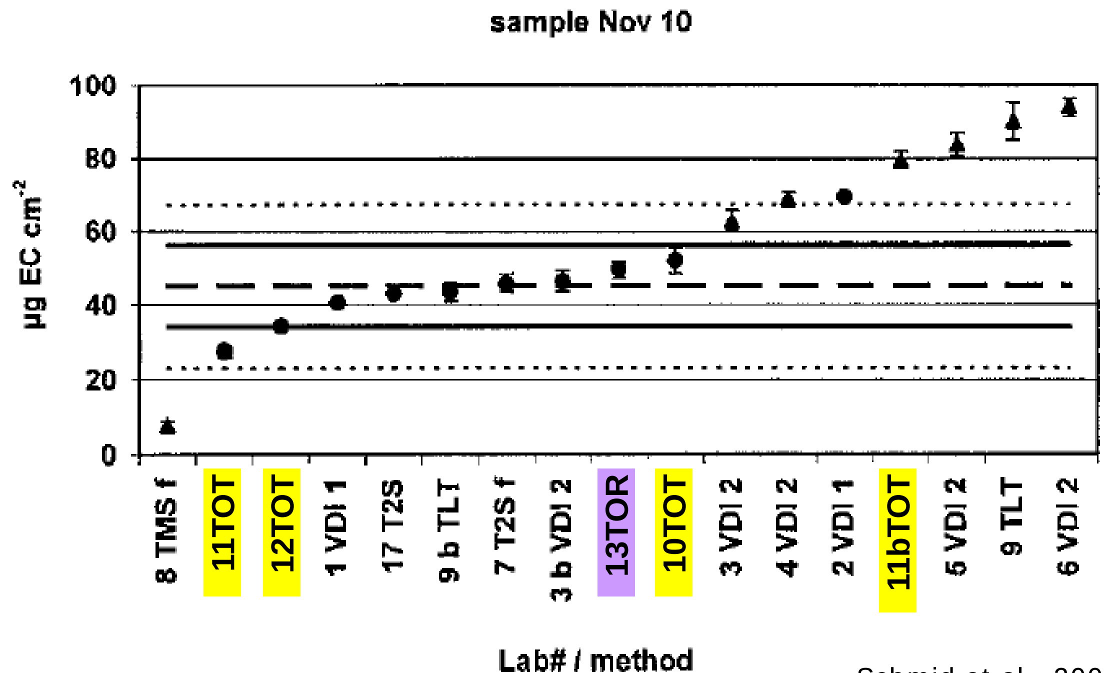
Chow et al., 2004

Different thermal evolution protocols give different results for elemental carbon





Schmid et al., 2001

California carbon emissions are dominated by biomass burning and engine exhaust (mostly diesel)

	2006 Emissions (tons/yr)		
Source Categories	**$PM_{2.5}$**	**BC**	**OC**
Fuel Combustion	11,727	1389	2268
Waste Disposal	230	18	42
Petroleum Production	781	34	68
Industrial Processes	17,178	231	4096
Solvent Evaporation	13	0	3
Miscellaneous (Wood stoves, fireplaces, waste)	164,111	12,609	48,381
Mobile (On-Road)	24,014	10,483	9703
Mobile (Other)	29,427	12,158	13,890
Wildfires	78,479[a]	15,161	29,530
Total	**325,959**	**52,084**	**107,979**

Chow et al., 2010

BC emissions can change by a factor of two with an equally justifiable source profile

Source Category	2006 BC Emissions (tons/yr) Base Case[a]	Based on CARB Gasoline and Diesel Profiles[b]	Based on Fresno RWC Profile[c]
Fuel combustion	1,389	1,135[c]	1,389
Industrial processes	231	231	231
Miscellaneous	12,609	12,609	10,684
Natural (wildfires)	15,161	15,161	15,161
Mobile (on-road)	10,483	5,876[c]	10,483
Mobile (other)	12,158	8,302[c]	12,158
Petroleum production	34	34	34
Waste disposal	18	18	18
Total	52,084	43,367[c]	49,885

Chow et al., 2010

Comparing different inventories can yield similar totals, but very different category emissions

Table 4. Comparison between 1995 California[a] and 1996 Bond et al.[30] black carbon (BC) and organic carbon (OC) emissions.

Emissions Inventory	Species	Base Year	Biofuel Combustion[b]	Fossil Fuel Combustion[c]	Open Biomass Burning[d]	Total
BC emissions in tons/yr (t/yr) (% of total)						
CARB[70]	BC	1995	4,741 (12%)	24,017 (62%)	9,978 (26%)	38,781
Bond et al.[30]	BC	1996	7,884 (24%)	16,109 (48%)	9,288 (28%)	33,281
OC in t/yr (% of total)						
CARB[70]	OC	1995	23,691 (29%)	30,332 (37%)	24,336 (30%)	82,596
Bond et al.[30]	OC	1996	42,991 (24%)	17,142 (10%)	118,968 (66%)	179,101
OC/BC ratios						
CARB[70]	OC/BC	1995	5.0	1.3	2.4	2.1
Bond et al.[30]	OC/BC	1996	5.4	1.1	12.8	5.4

Notes: [a]California Air Resources Board (CARB) emission inventory: http://www.arb.ca.gov/app/emsinv/fcemssumcat2007.php. [b]Biofuel burning includes residential wood combustion (RWC) and cooking. [c]Fossil fuel combustion includes fuel combustion, industrial activities, petroleum production, miscellaneous processes, and on-road and other mobile sources. [d]Open burning includes agricultural waste burning, forest management, and wildfires.

Chow et al., 2010

Improving and using California BC inventories

- Incorporate BC measurements into certification tests and ensure compatibility with ambient measurement methods (CARB, 2011). http://www.arb.ca.gov/regact/2012/leviiighg2012/levappp.pdf
- Create a push system for accumulating and describing profile measurements
- Separate emissions for the flaming and smoldering phases of biomass burning
- Quantify spectral absorption properties of source and ambient samples
- Incorporate carbon emissions into annual emission summaries

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