Where There's Smoke There's Haze:

Estimating the Contribution of Fire Types to PM2.5 and Haze

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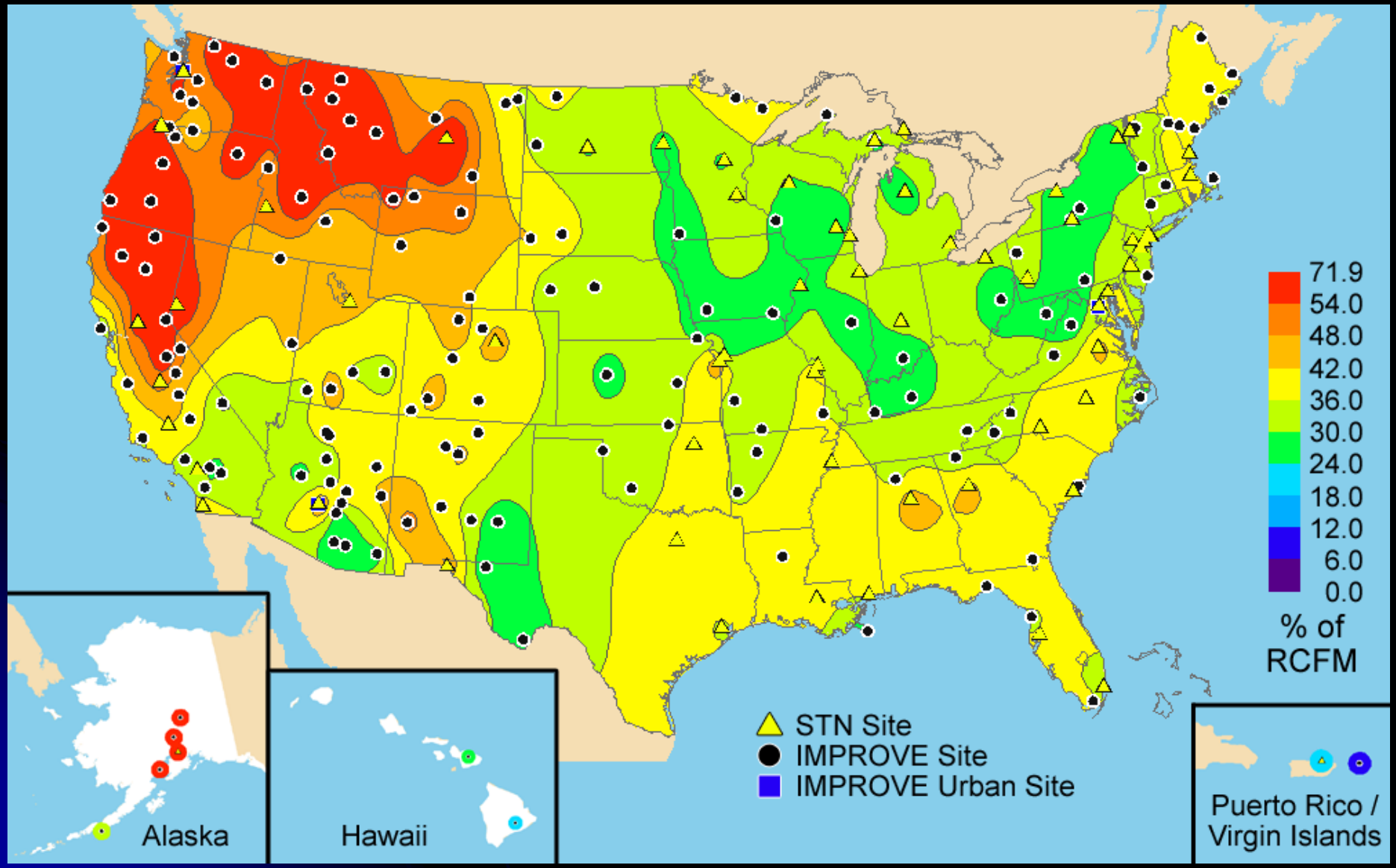
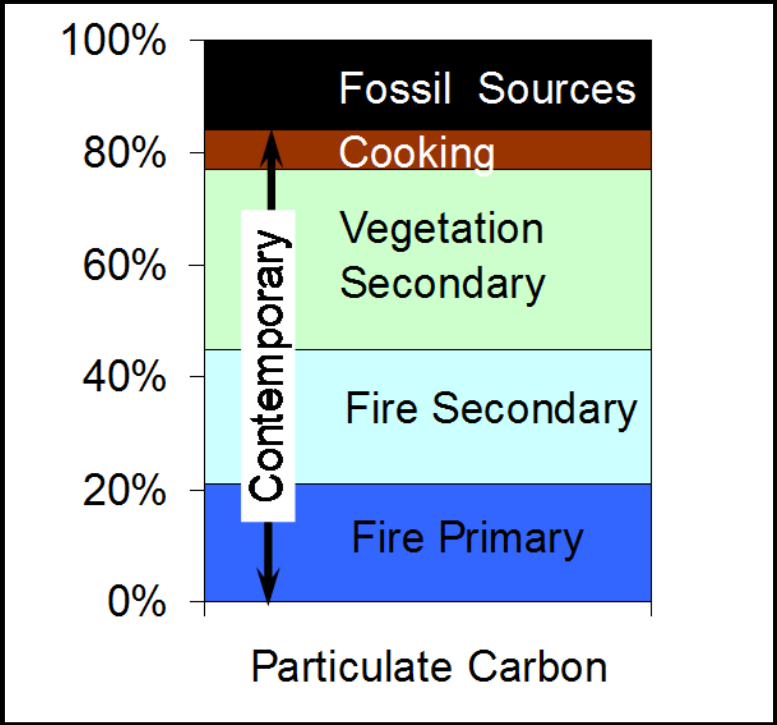
Urban & Rural Average Annual Organic Carbon PM2.5 Fraction

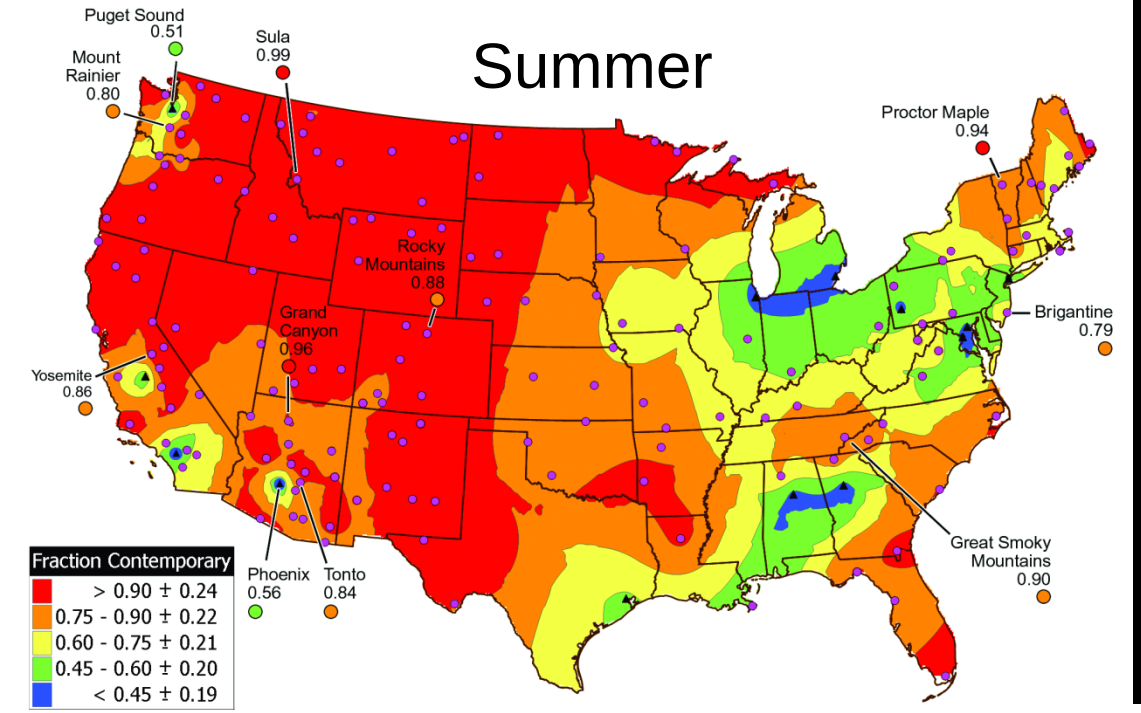


Sources Contributing to Particulate Carbon

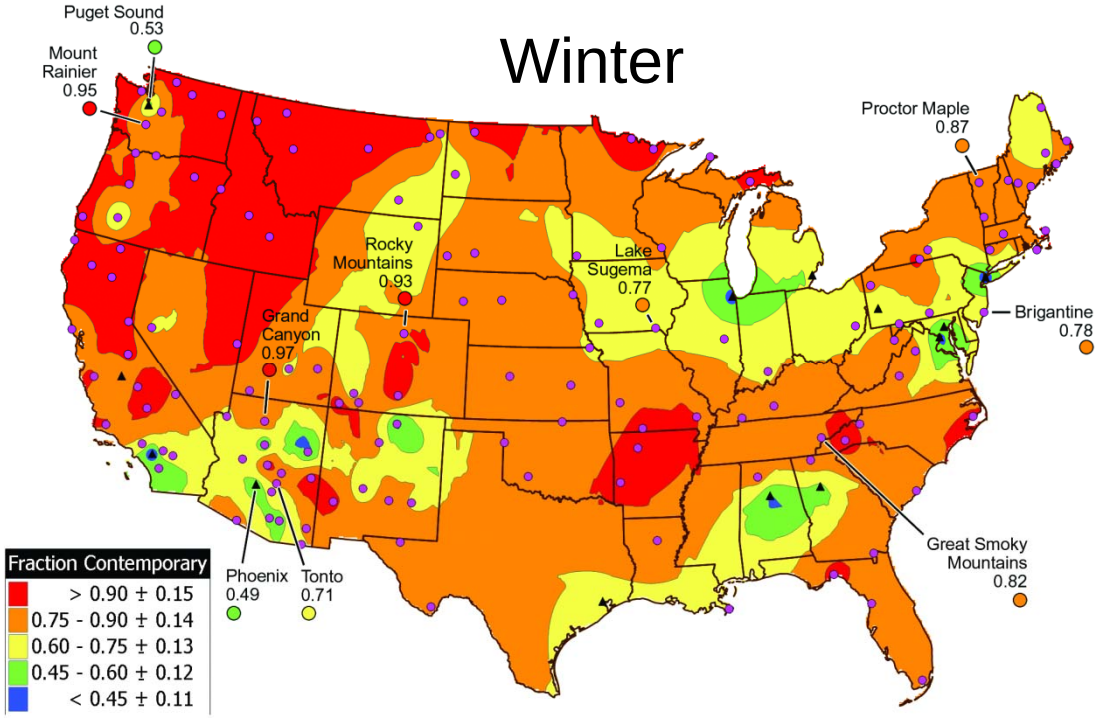


- Source can be divided into a contemporary or fossil fraction
- Contemporary or biogenic carbon source include
  - Fires, SOC from vegetation, cooking, pollen, and others
- Fossil or “old” carbon arises from burning of fossil fuels
- A large fraction of ambient particulate carbon is secondary organic carbon (SOC) formed from emitted organic gases

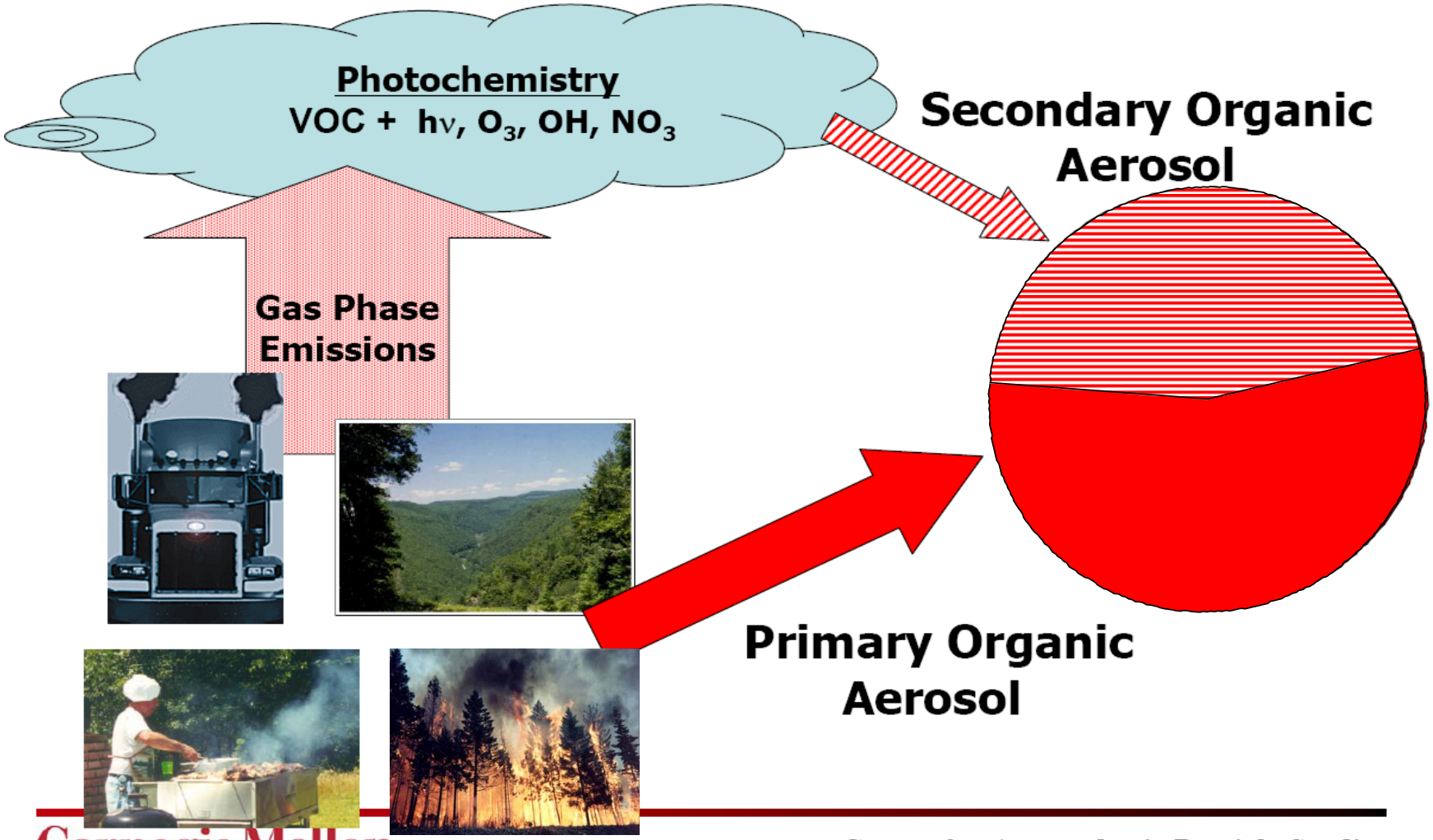
Radiocarbon (^{14}C) Distinguishing Between Contemporary and Fossil Carbon



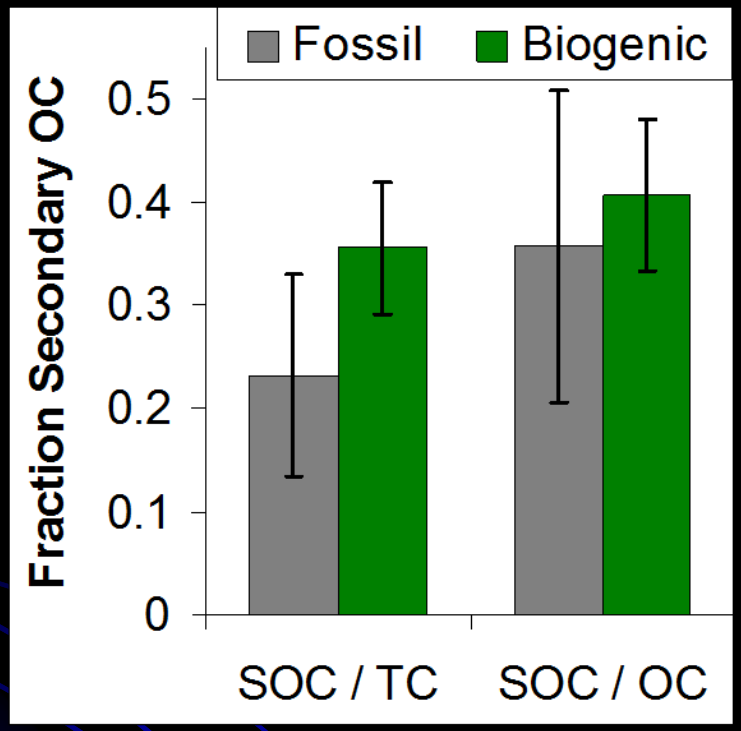
- Fraction Contemporary C
  - 80-100% - rural sites
  - 70-80% - near urban sites
  - 50% - urban sites
  - 60-75% in industrial Midwest
- Similar fraction contemporary carbon in winter and summer



Sources of Organic Aerosol (OA)



Summertime Fraction of SOA in Fossil and Contemporary Carbon

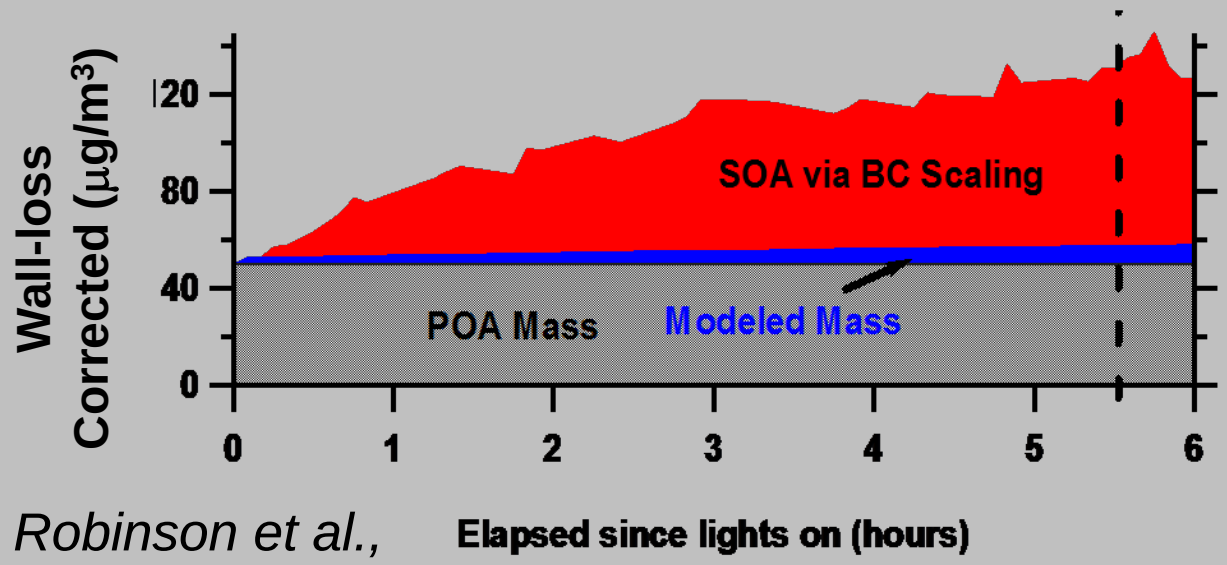


	Secondary TC	Secondary OC
Biogenic	36% (6.4)	41% (7.3)
Fossil	23% (10)	36% (15)

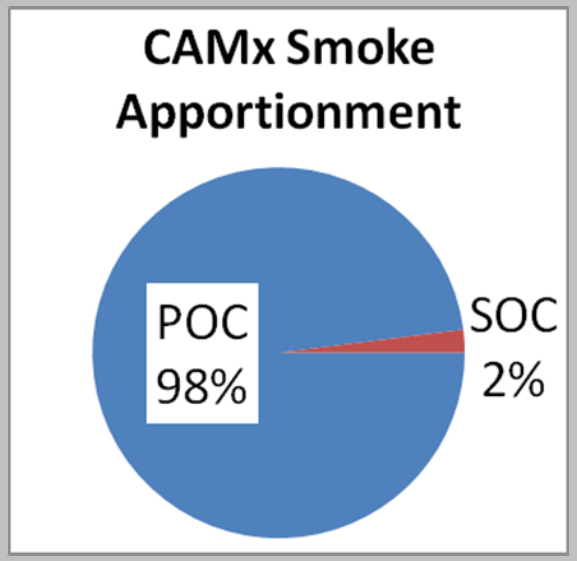
- Assumes all winter organic carbon is primary
  - Underestimates the summer secondary particulate carbon
- Assumes that a similar mix of sources contribute to the particulate carbon in the summer and winter.
  - Impact on estimate is unknown

SOA from Biomass Emissions

Laboratory and field study have shown evidence for large contributions of SOA



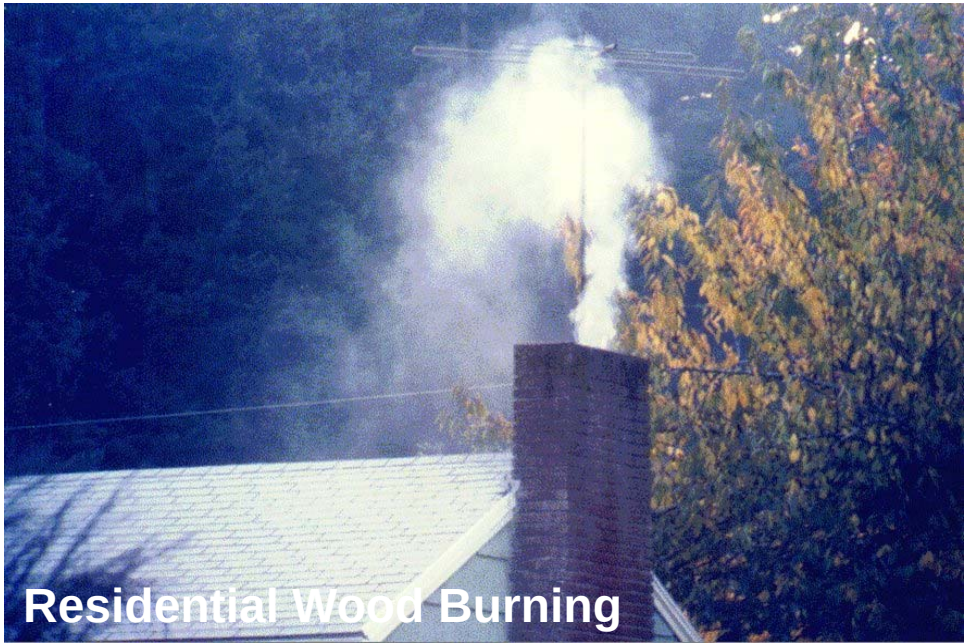
Robinson et al.,



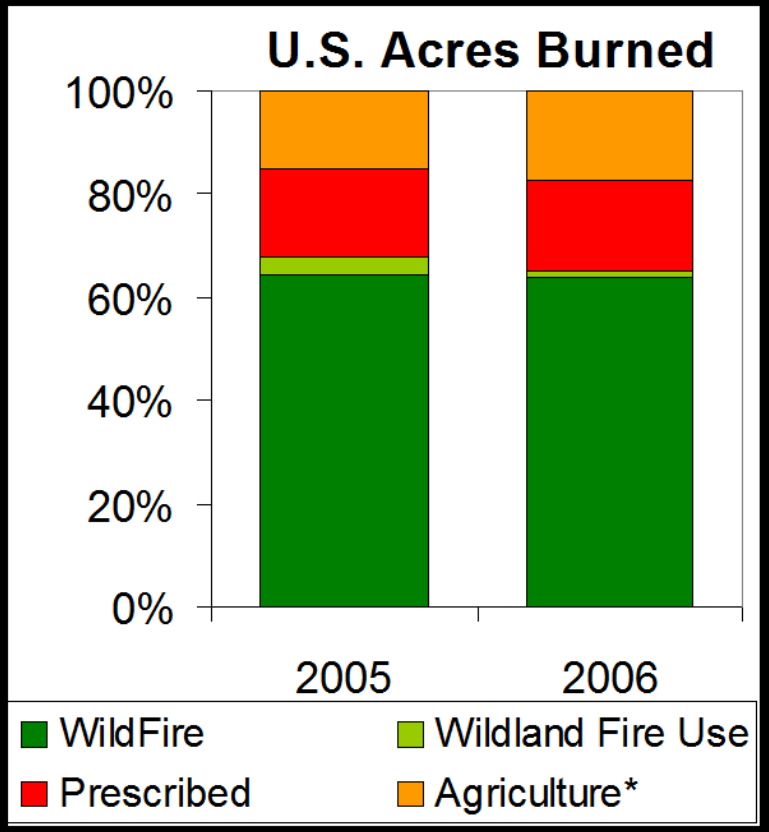
Aging of VOCs from biomass burning rapidly creates a lot of SOA, doubling the organic carbon concentrations

Current state of the art modeling estimates little to no SOA from biomass burning

Contribution of Fires to Particulate Carbon

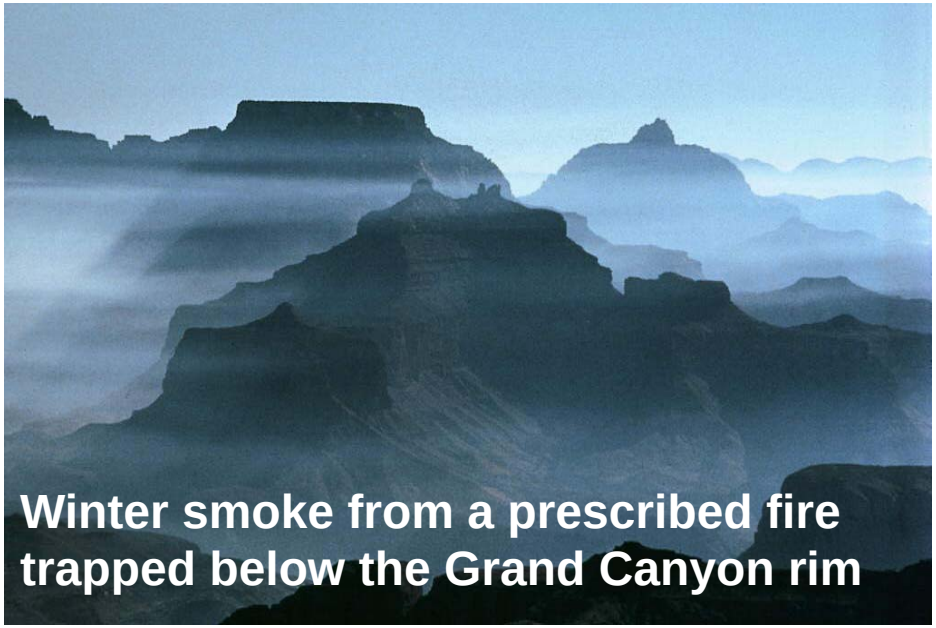


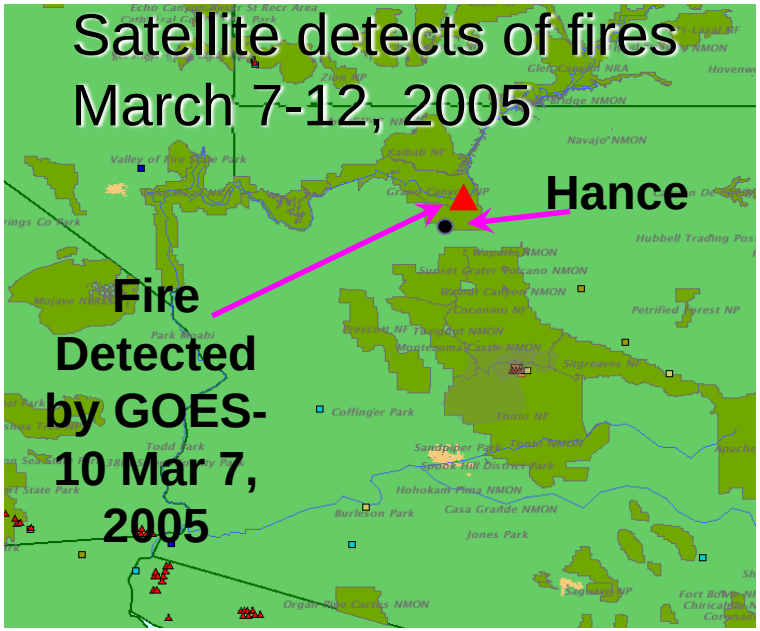
Emissions from Different Fire Types

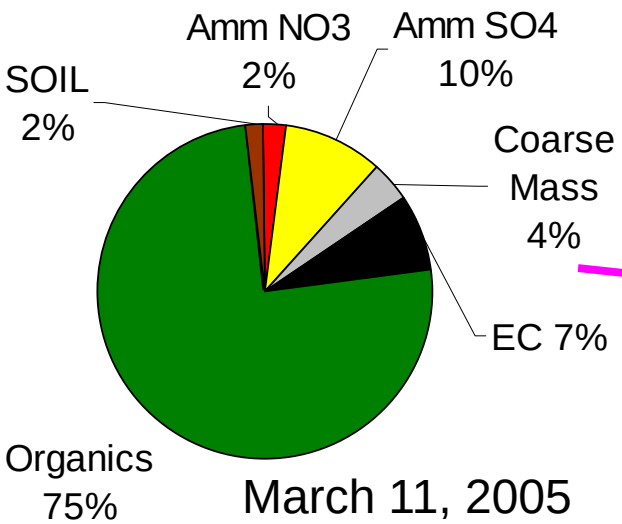


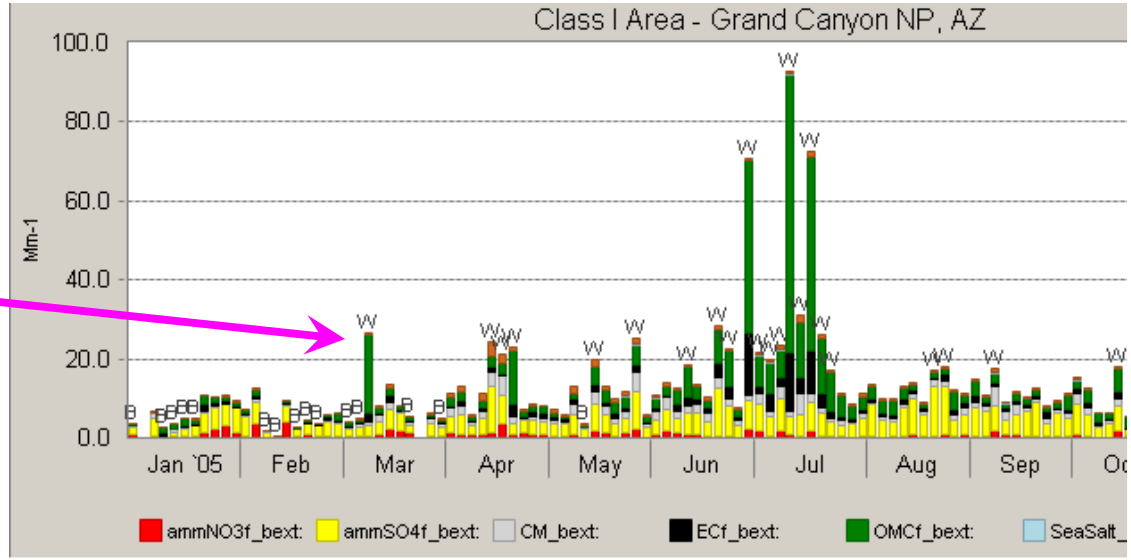
- Wildfire and wildland fire use, “Natural fires”, are the largest sources of smoke, especially in the western United States accounting for >80% of the acres burned.

Prescribed Fire in the Grand Canyon

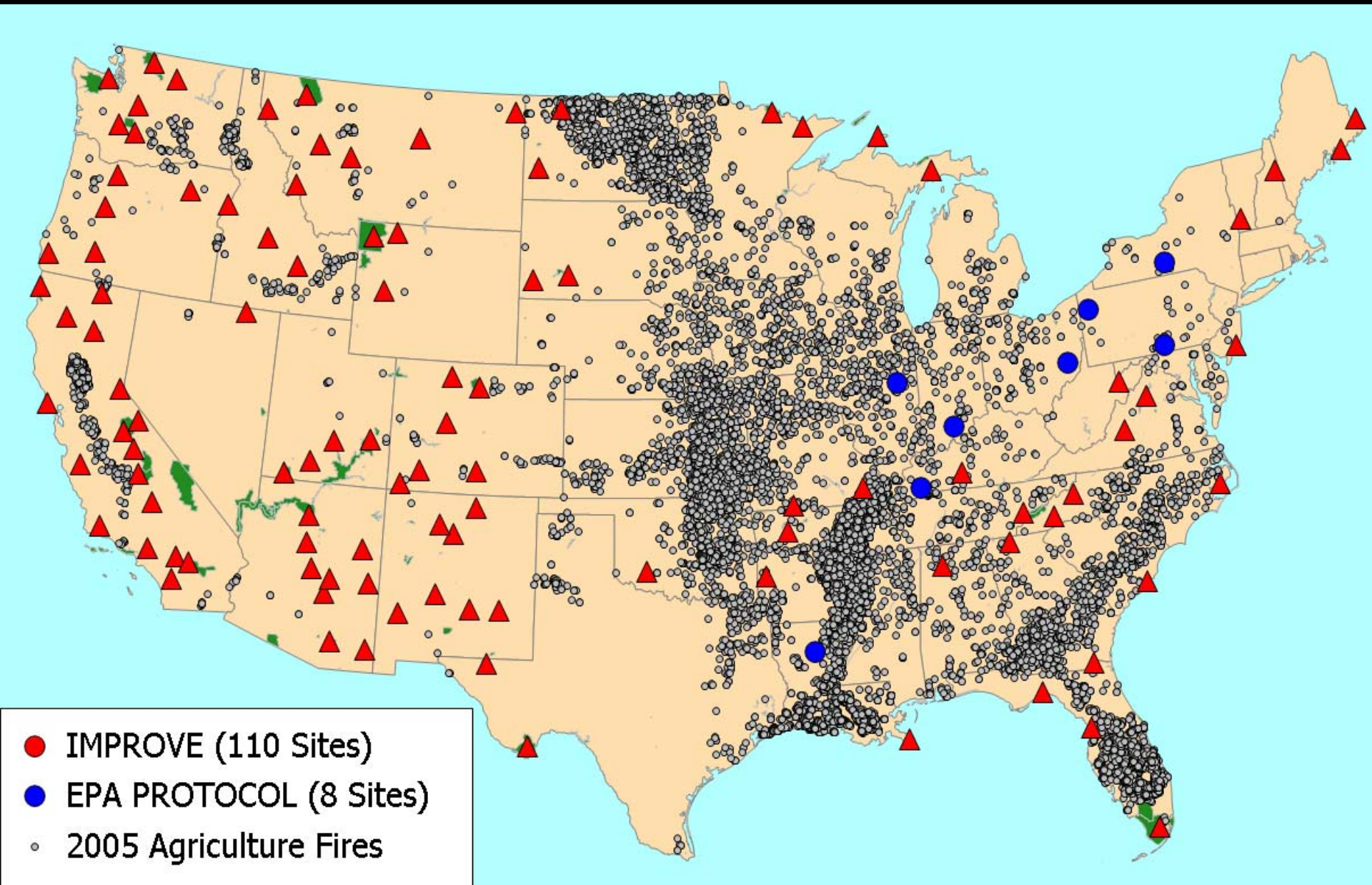








2005 Agricultural Fires

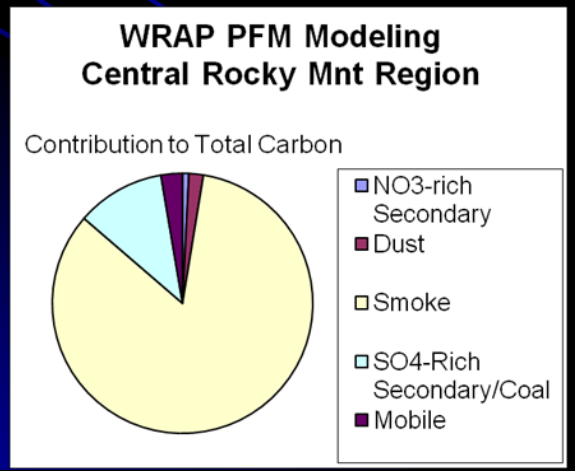


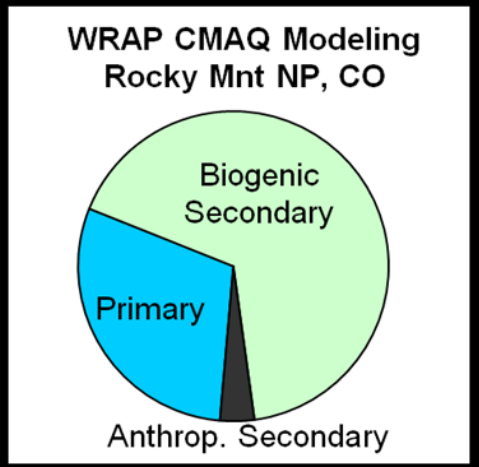
Smoke Management Needs for Air Quality Regulations

- Develop an unambiguous routine and cost effective methodology for apportioning primary and secondary carbonaceous compounds in PM2.5 RETROSPECTIVELY to prescribed, wildfire, agricultural fire, and residential wood burning activities
  - Daily contributions needed for Haze Rule to properly estimate natural contribution and contribution to worst 20% haze days
  - Annual and daily contributions needed for PM2.5 and PM10 NAAQS
  - Long term data needed to assess successes of smoke management policies
- Similar needs for ozone and reactive nitrogen deposition issues

Apportionment Methods

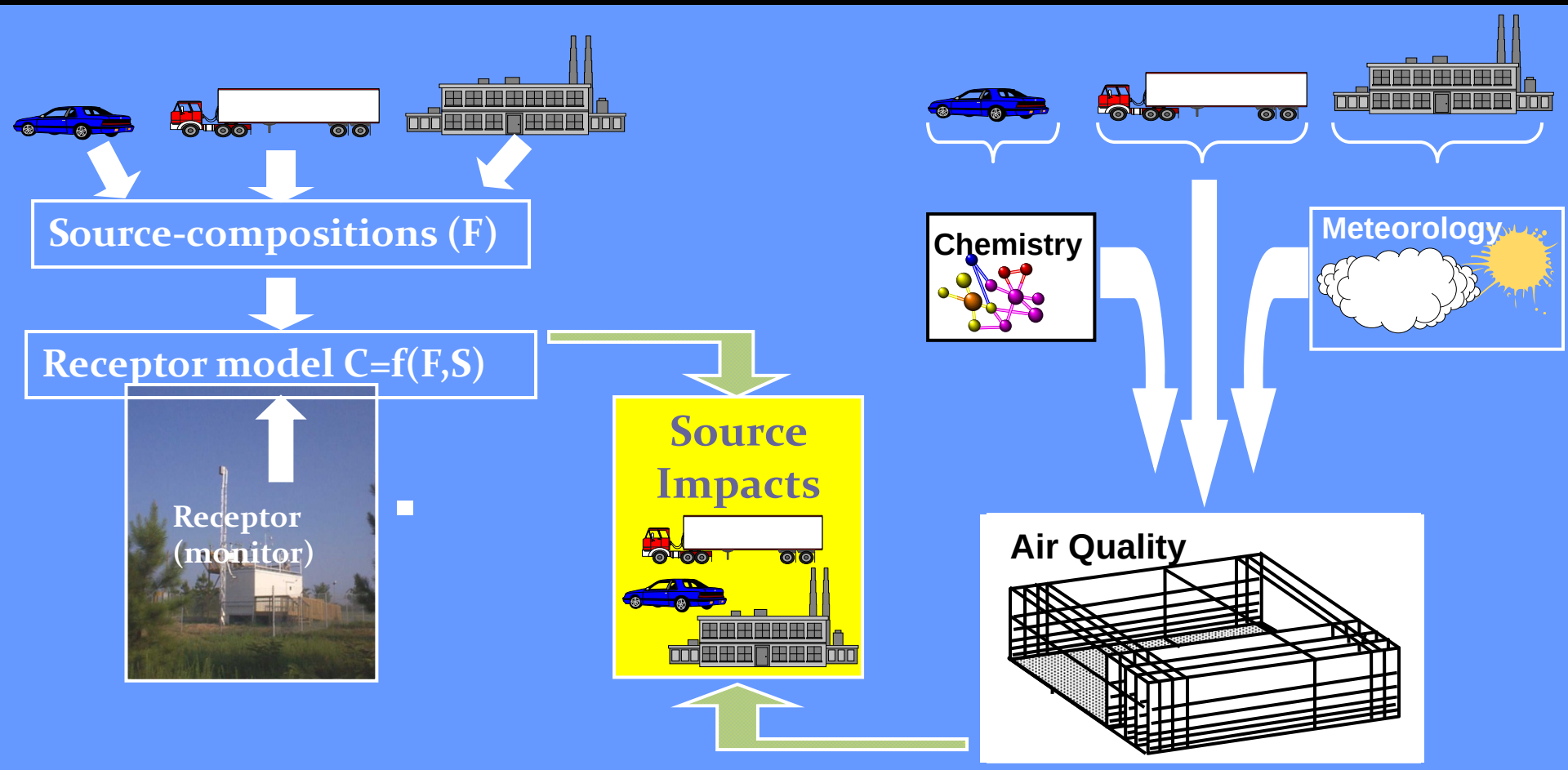
- Chemical transport models CTM)
  - Subject to large errors in inputs
    - Current CTM model simulations estimate little to no smoke SOA
  - Results are unconstrained by measured data
- Receptor Models
  - Chemical Mass Balance
    - Can't apportion secondary aerosols
  - Factor Analysis e.g. Positive Matrix Factorization (PMF)
    - Non-unique source factors





PMF modeling appears to have combined SOC from vegetation in with fire

Hybrid Source Apportionment Model

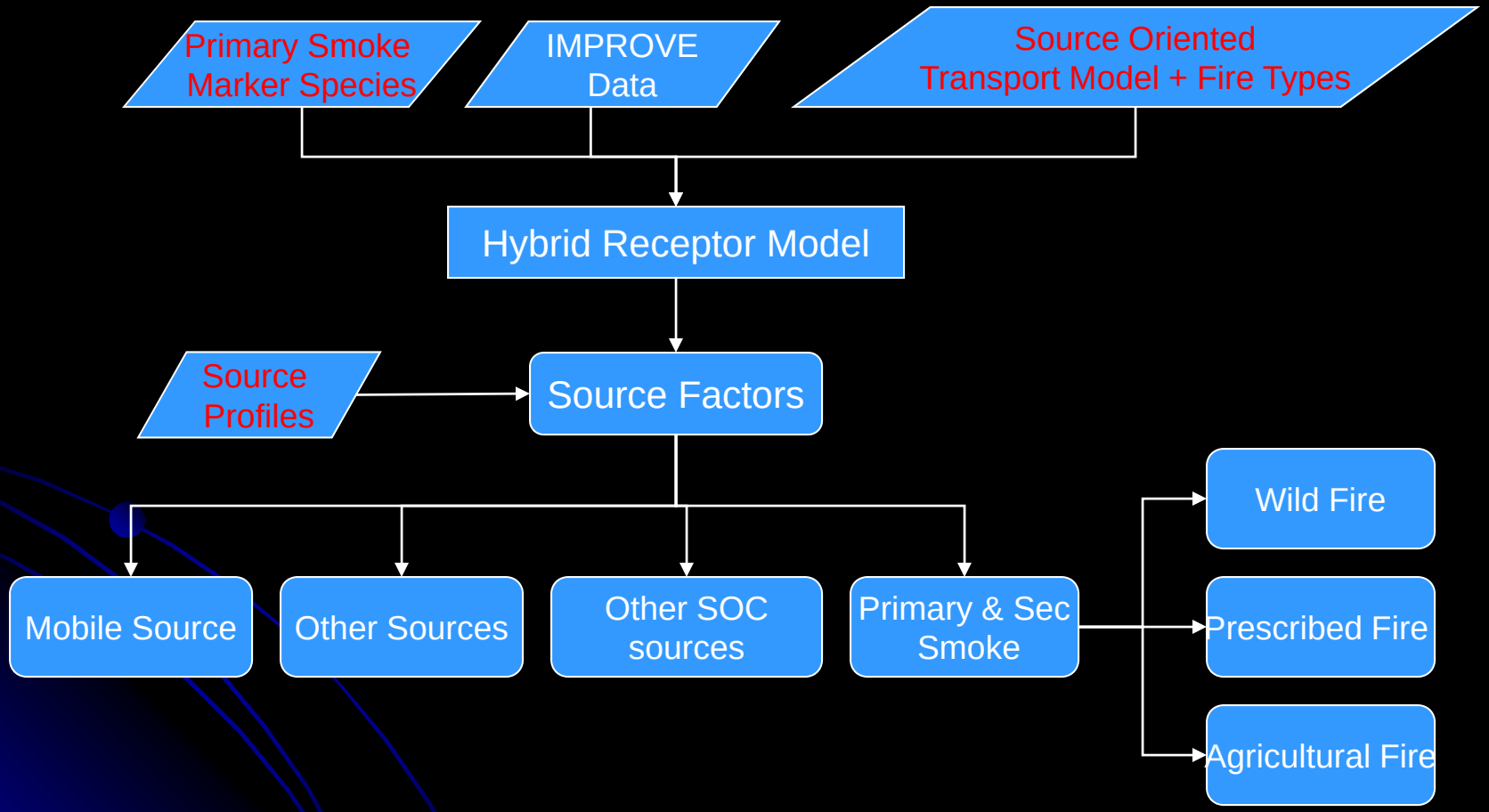


Receptor Model
(CMB, PMF)

Source-oriented Model
(3D Air-quality Model)
(CMAQ, CAMx)

Jeameen Baek et al., - Georgia Institute of Technology

Smoke Apportion: Hybrid Receptor Modeling



Developing a Retrospective Smoke Apportionment System

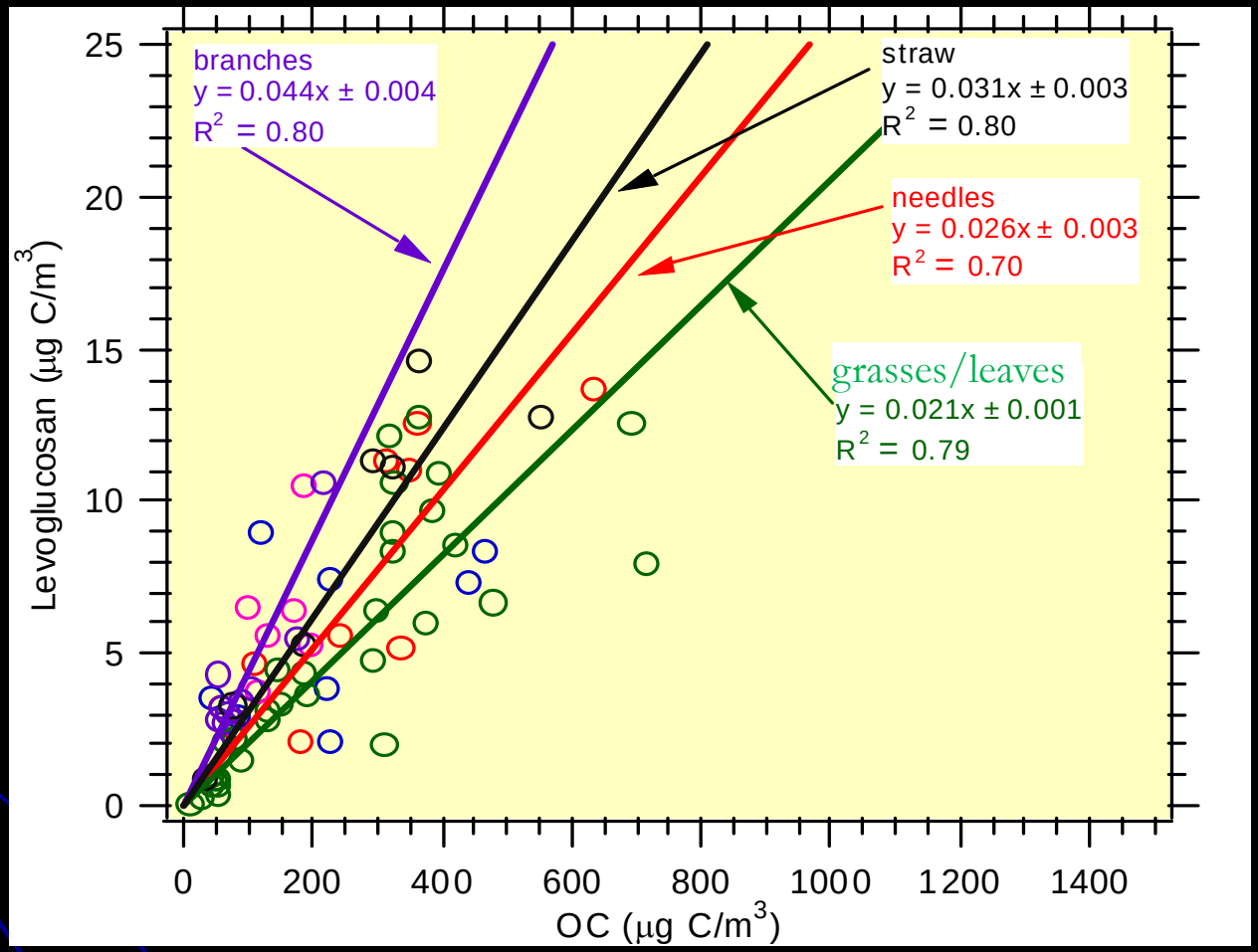
- Source apportionment system to estimate the contribution of primary and secondary smoke from different types of fire
  - Primary Smoke
    - Cheap and easy smoke markers species (Levoglucosan) measurements methods applicable in routine monitoring programs
    - Smoke source profiles for wildland fuel types
  - Secondary Smoke and Smoke Types
    - Hybrid source apportionment model - Statistical model for integrating deterministic modeling results and measured data

The FLAME Experiment

- USDA Forest Service Fire Science Lab at Missoula
- Characterization of primary smoke emissions
  - Hundreds of burns
  - Fuel components and complexes
  - NW, SW, and SE fuel emphasis
  - Chemistry, optical properties, hygroscopicity

Levoglucosan vs. OC

- branches
- straw
- needles
- leaves
- grasses
- duffs



-Levoglucosan is unique to fire and stable in the atmosphere

-Stable ratio's for different fuel types

-In a lab fire levoglucosan is a very good smoke marker species

Hybrid Receptor Model Incorporating Prior Source Attribution Estimates in Chemical Mass Balance Eq

$$\left[x_{ij}, y_{il}\right] = \sum_{k=1}^{p} g_{ik}\left[f_{kj}, b_{kl}\right] + \left[e_{ij}, e^{y}{}_{il}\right]$$

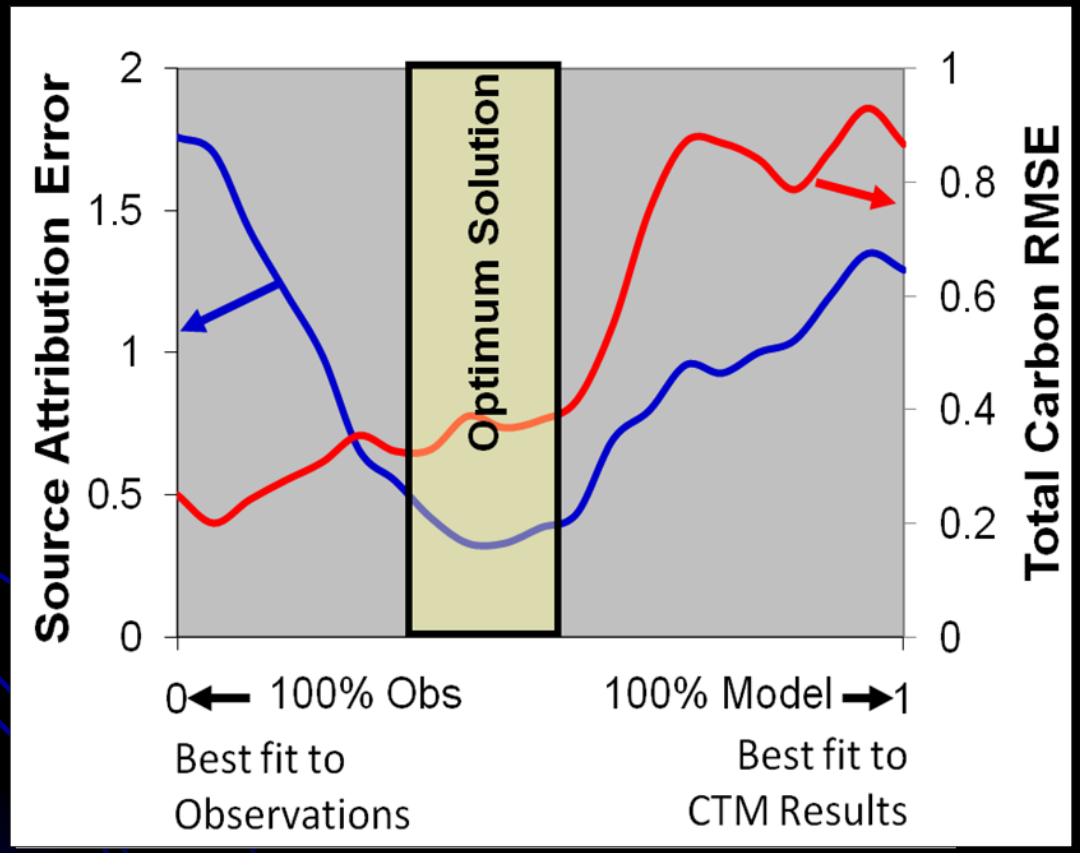
- $y_{il} = g_{ik} * b_{kl} \pm e^{y}_{il}$ - Prior source contributions from the kth source to the ith measurement at the receptor.
- - Multiplicative coefficients relating prior source attributions estimates, ***Y*** to **G**
- Positive Matrix Factorization (PMF):
  - Only the ***X*** and ***Y*** are known and the CMB equation is solved for the source contributions (***G***), source factors (***F***) and ***b***

Incorporate CTM results into PMF optimization technique

$$Q = \sum_j \sum_i \frac{\left(x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}\right)^2}{s_{ij}^2} + \sum_k \sum_i \frac{\left(y_{ik} - \sum_{l=1}^{p} g_{ik} b_{lk}\right)^2}{{}^{y}s_{ik}^2 / \lambda}$$

- s_{ij} – uncertainty in the jth species for measurement I
- ${}^{y}s_{ik}$- uncertainty of the kth source type for CTM modeled period i
- It is assumed that ${}^{y}s_{ik}$ are known within some constant λ
  - By modifying λ the relative weight of the measured and modeled data on the solution can be adjusted. Therefore λ can act as a tuning coefficient
- **G**, **F** and **B** are found by minimizing Q using the constrained weighted least square method in PMF

Hybrid Model – Trade-offs between reproducing the measured and modeled data

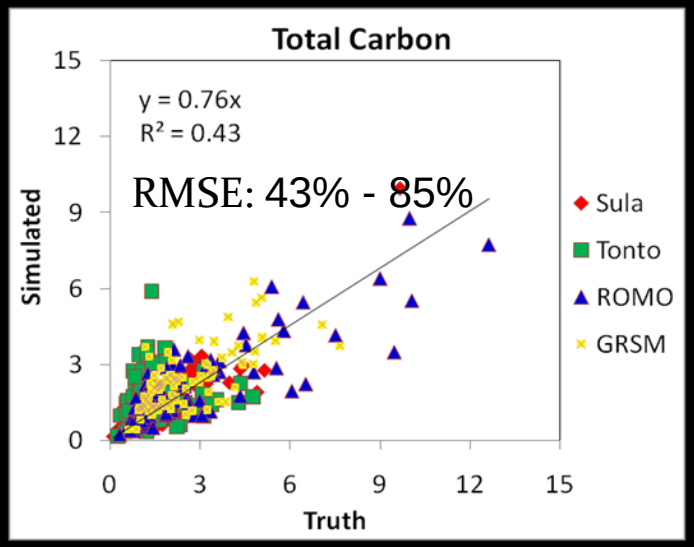


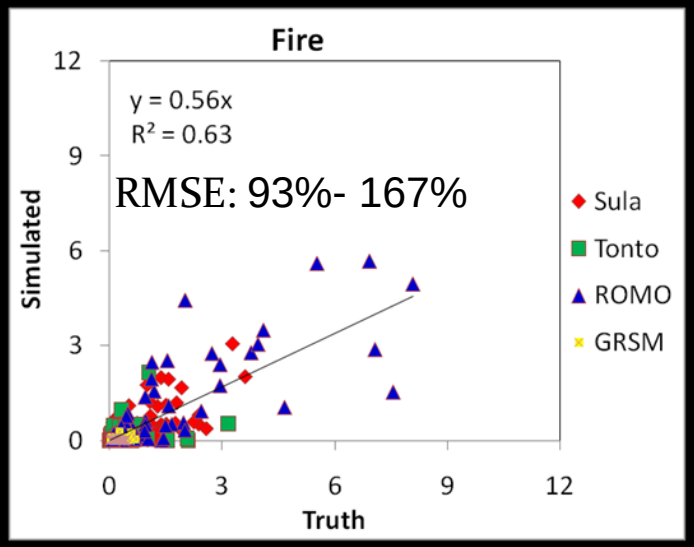
increasing λ

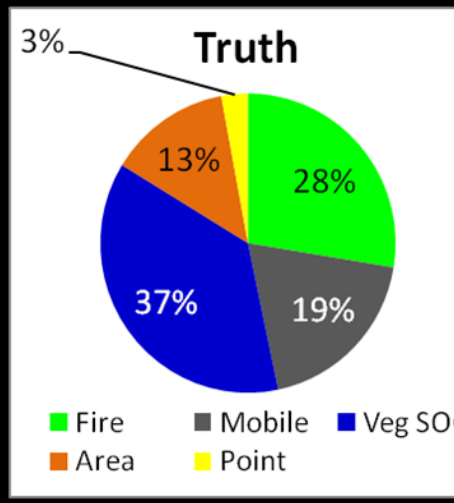
Testing the Hybrid Model using Synthetic Data

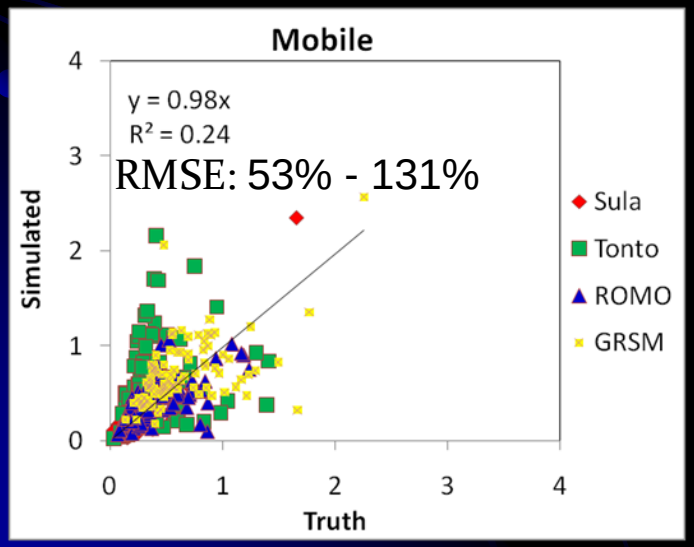
- **True Source Contributions**
- **Prior Source Contribution estimate (CTM modeling results)**
- **Synthetic Measurements**

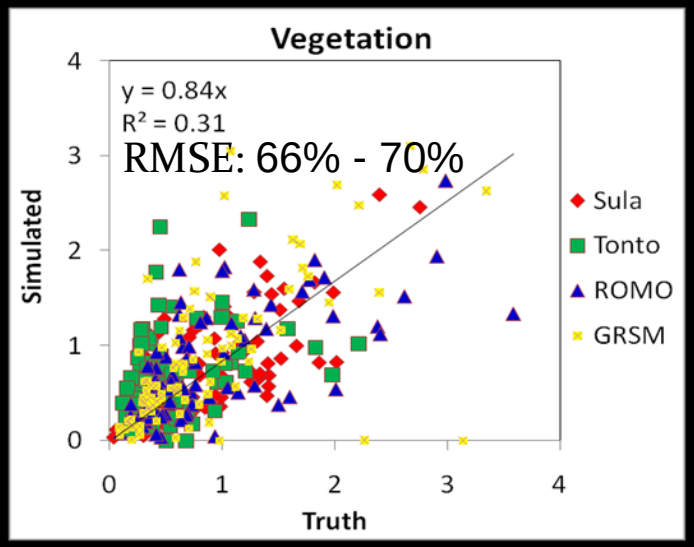
Source Contributions: Truth vs. Prior Estimates

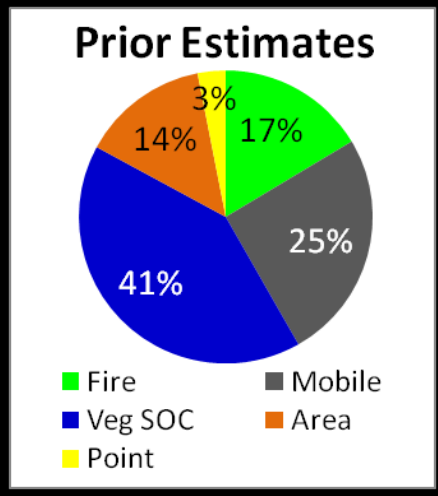




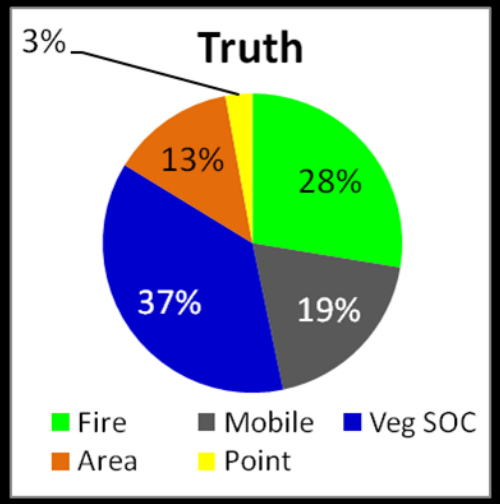


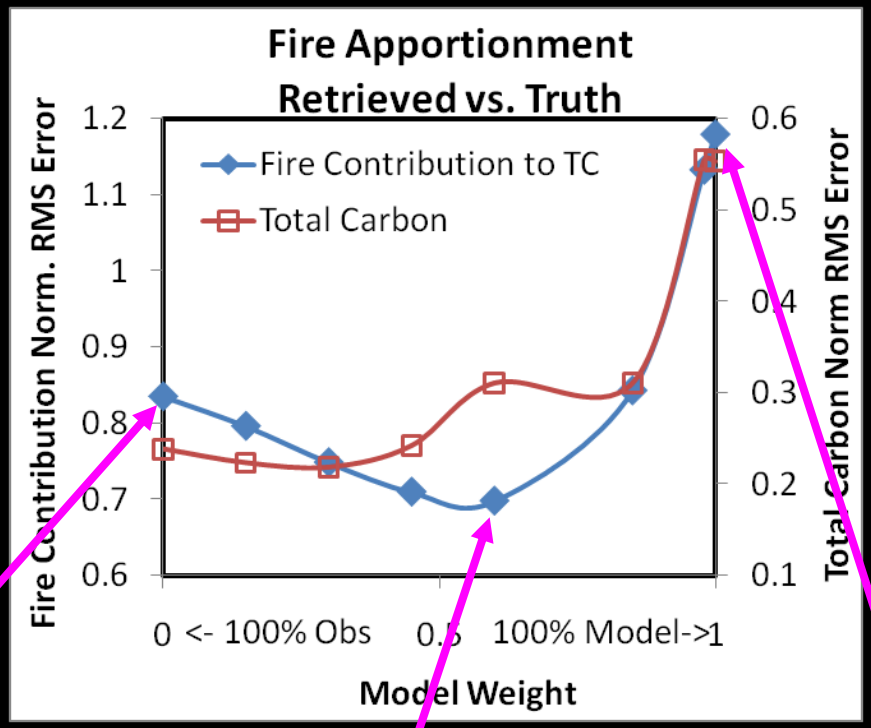


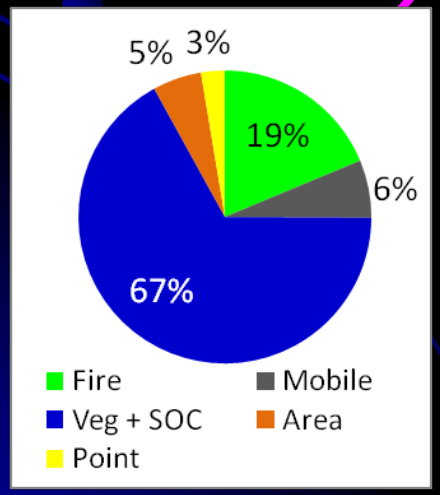


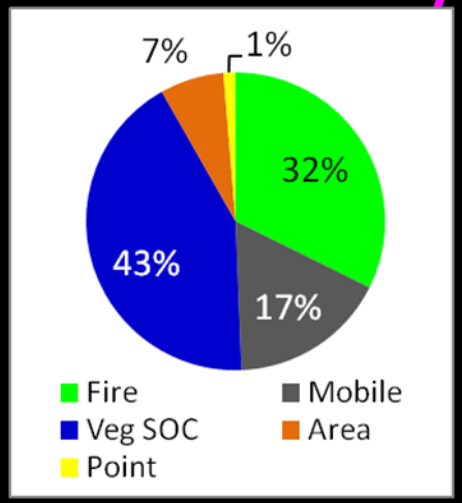


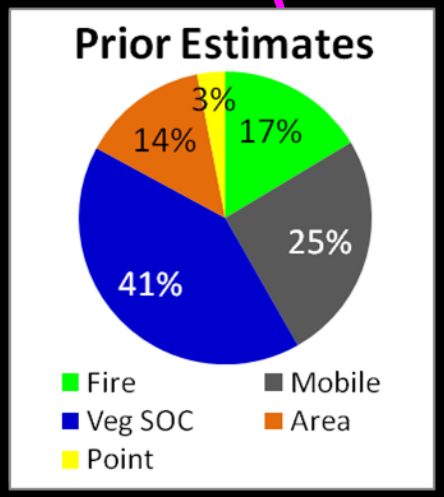
Hybrid - PMF Results





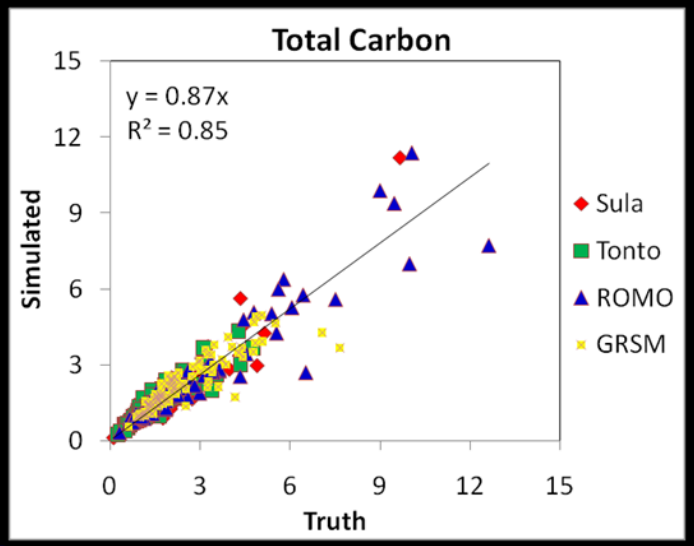


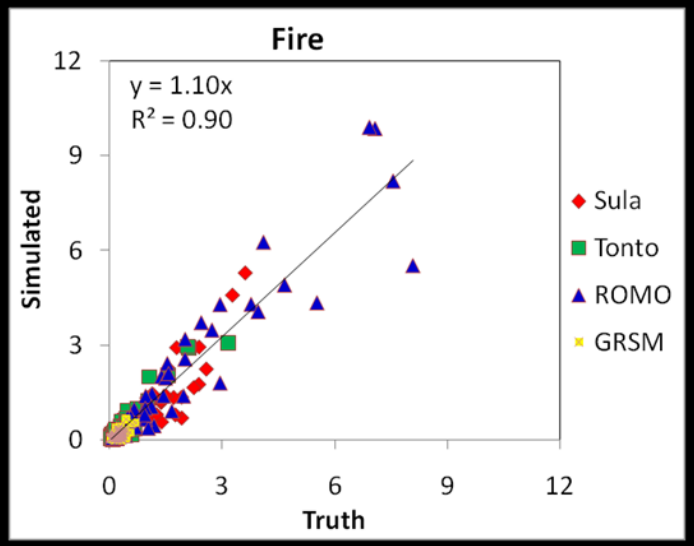


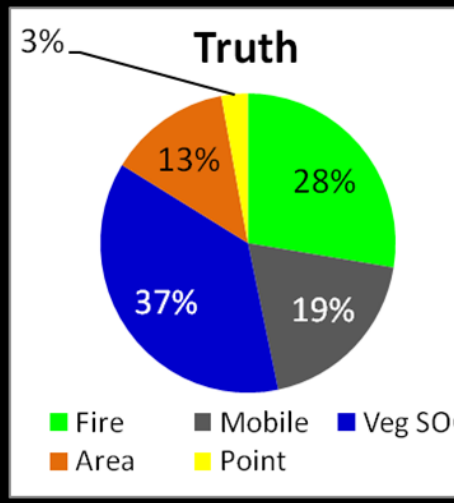


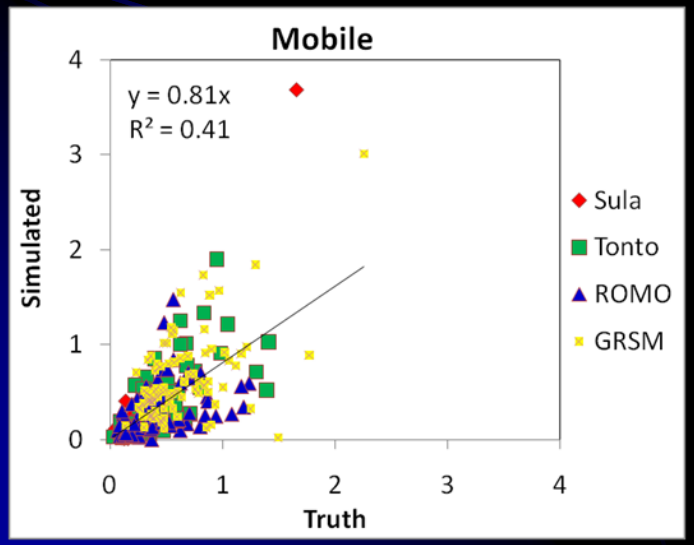
Hybrid Model vs. Truth

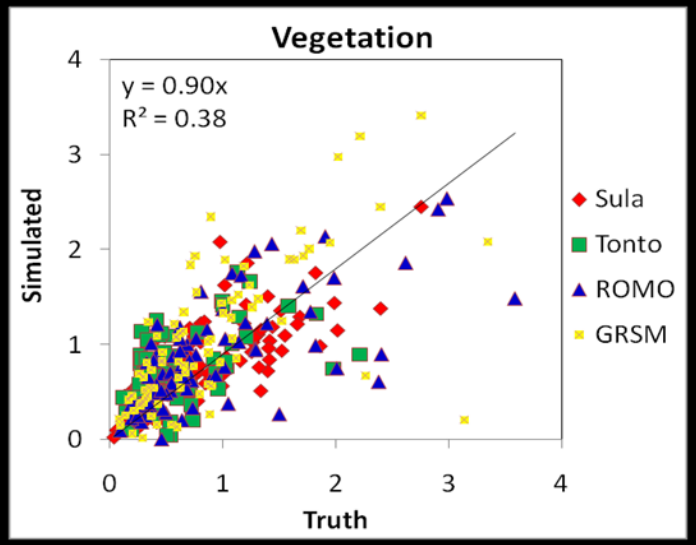
Model and Observation have near equal weight

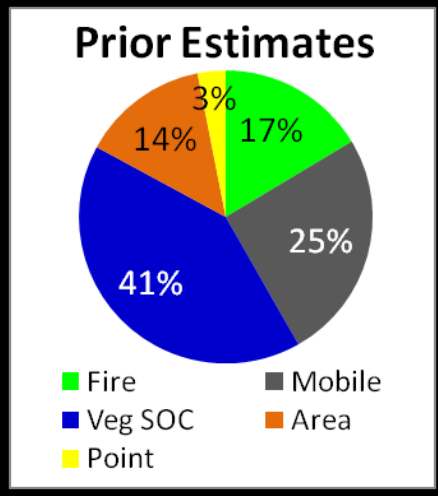












Routine Data Needs for the Hybrid Receptor Model

- Measured aerosol data – IMPROVE monitoring network
- Smoke marker species – not routinely measured
- Prior smoke and other source attribution results
  - Will generate daily source attribution estimates for CIA using simply trajectory based CTM results
- Fire emissions tagged by source types
  - WRAP FETS is collecting the needed information to do this

Further Developments Needed

- Hybrid model capable of estimating contributions from different fire types
- Field studies to validate and further develop smoke marker species source profiles
- Further testing of the hybrid model on real data

- Blue Sky modeling framework is a logical place for implementing a hybrid receptor model