

Historical Context of the SOAS 2013
Centreville Field Study:
Impacts of Emissions on PM_{2.5} EC and OC
in the SEARCH Network, 1999 - 2013

SAS Data Analysis Workshop

March 31 – April 2, 2014

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SEARCH

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ENVAIR

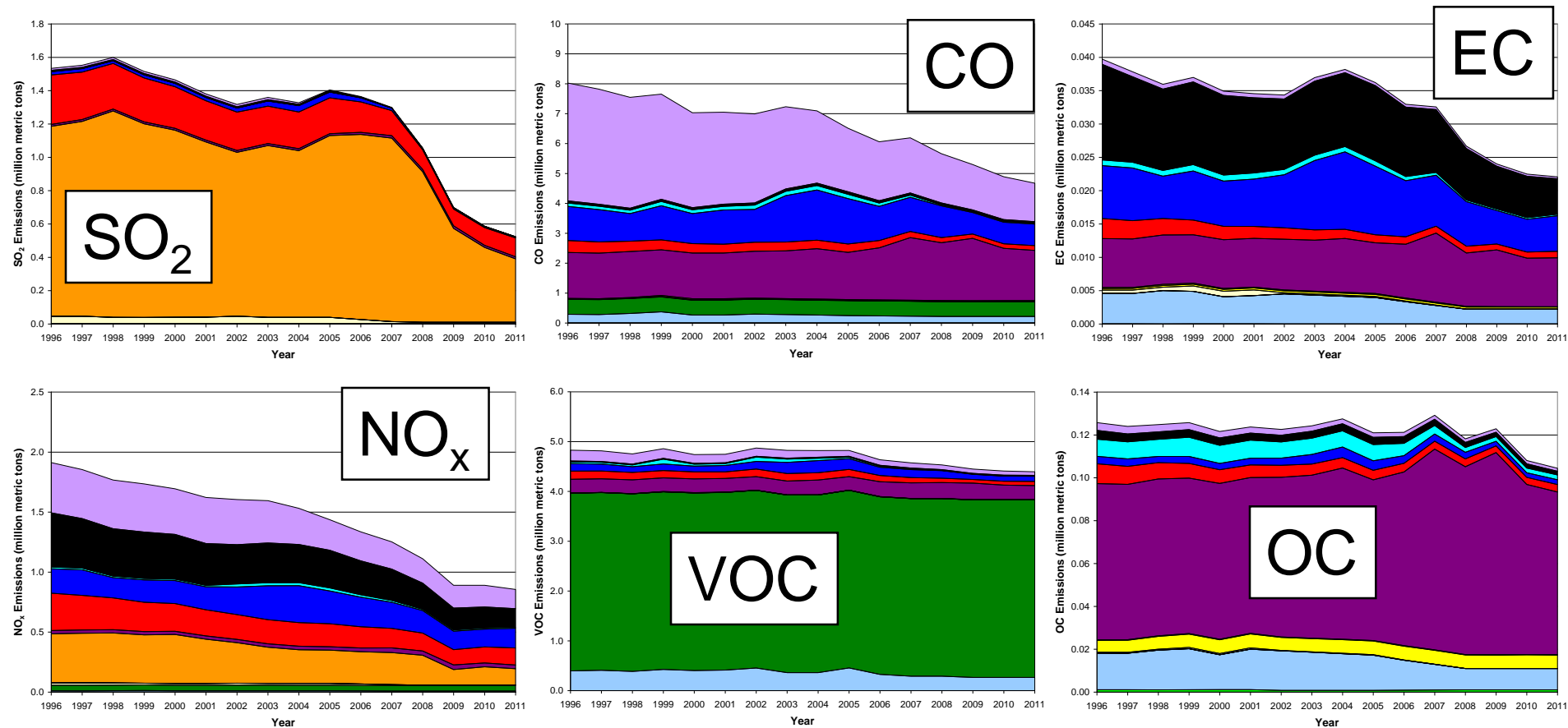
Question

Were urban & rural SEARCH sites substantially impacted by mobile-source emissions and biomass burning between 1999 and 2013?

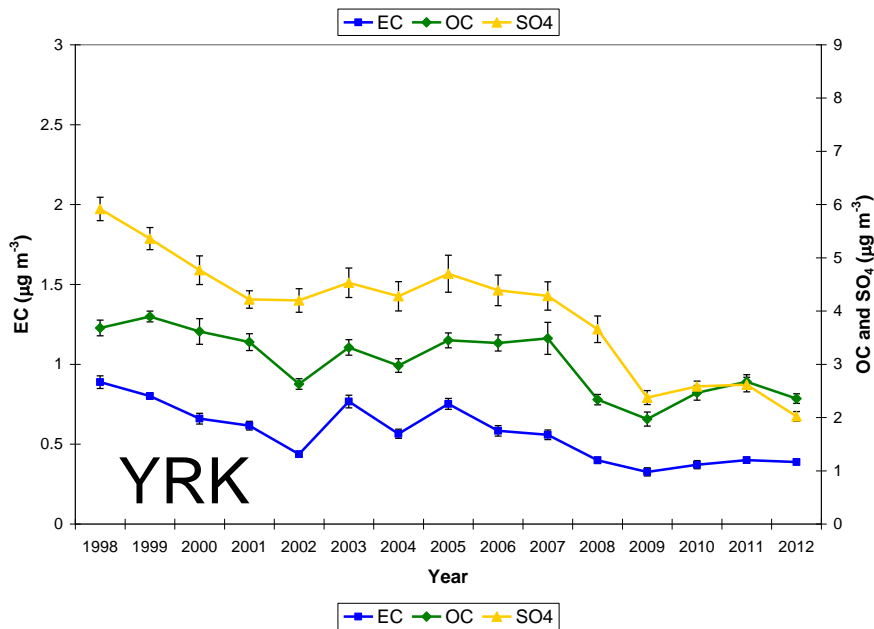
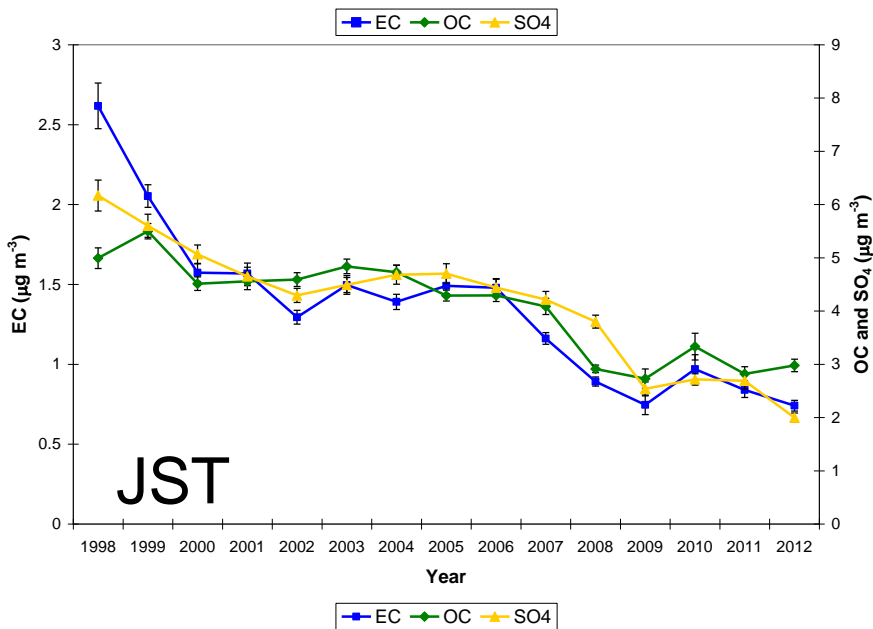
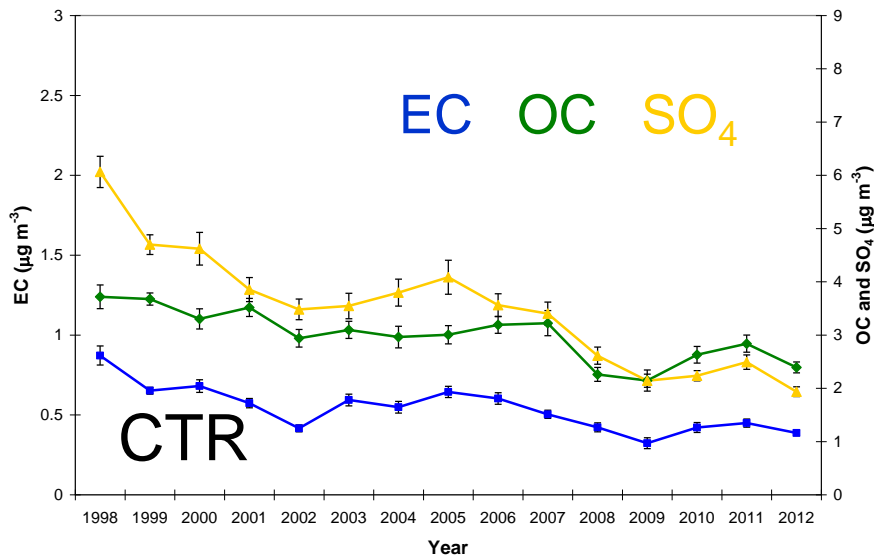
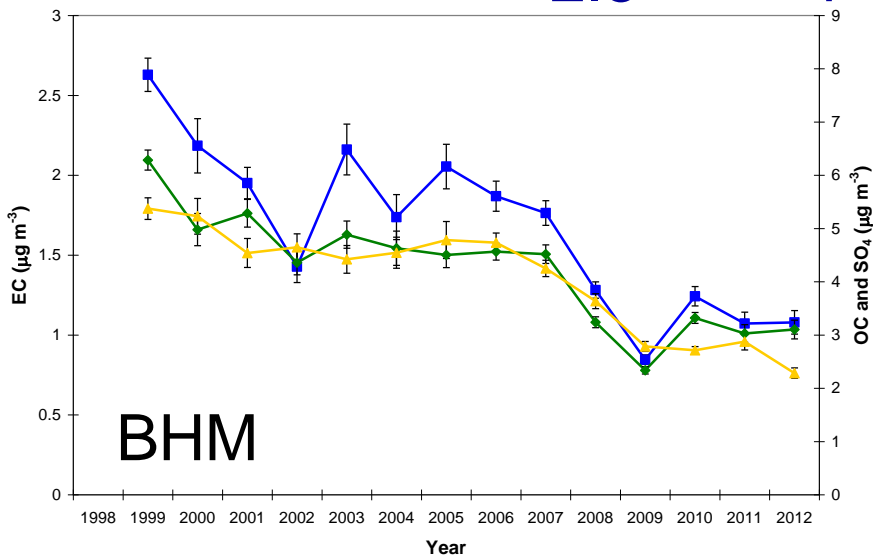


What are the magnitudes of the impacts?

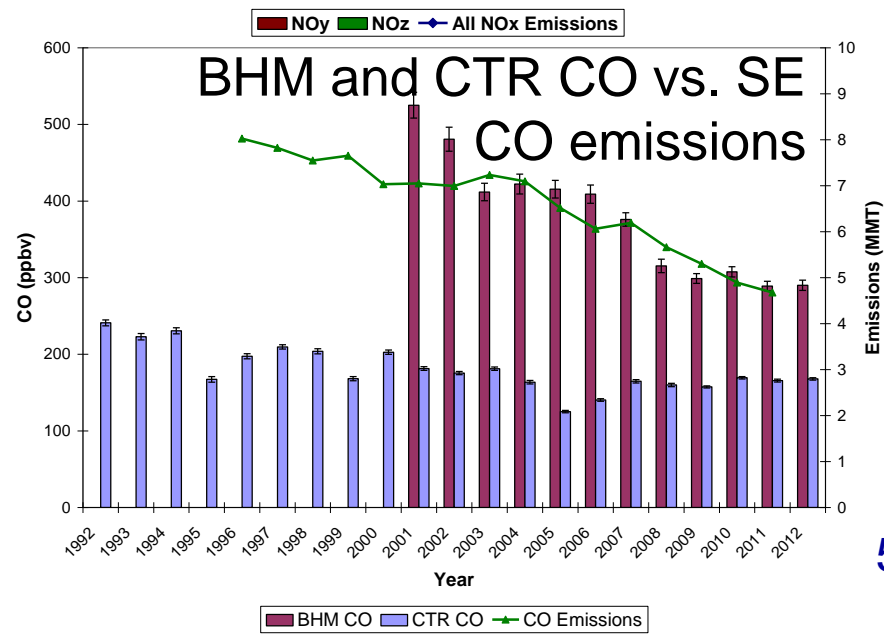
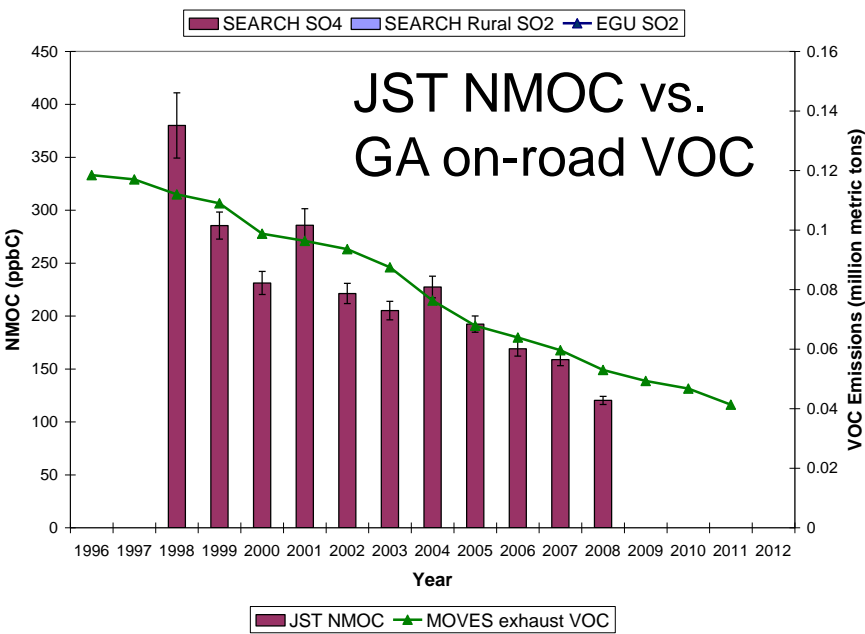
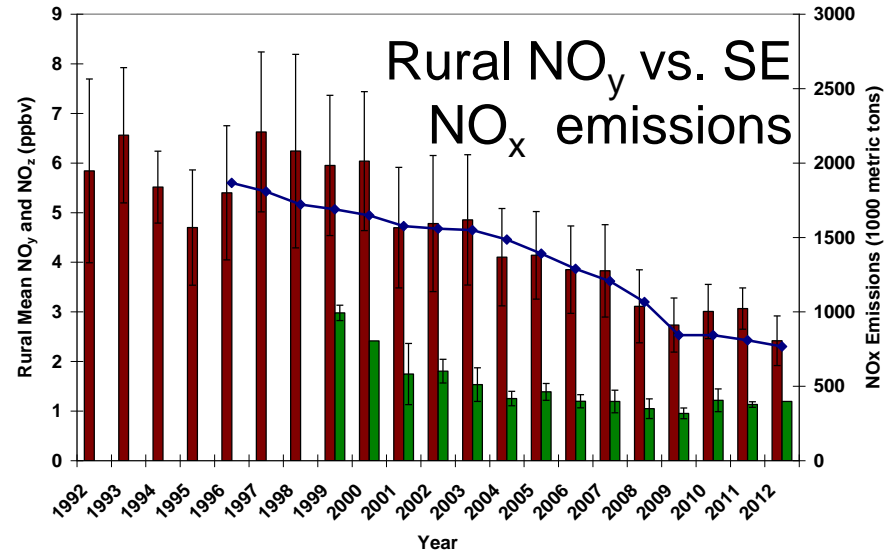
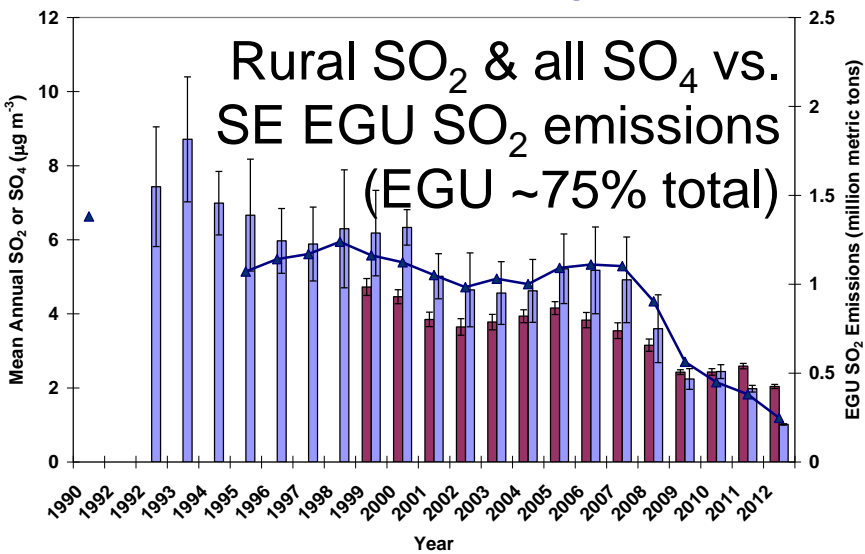
Decreasing Anthropogenic Emissions in AL, GA, MS, NW FL (SE), 1996 - 2011



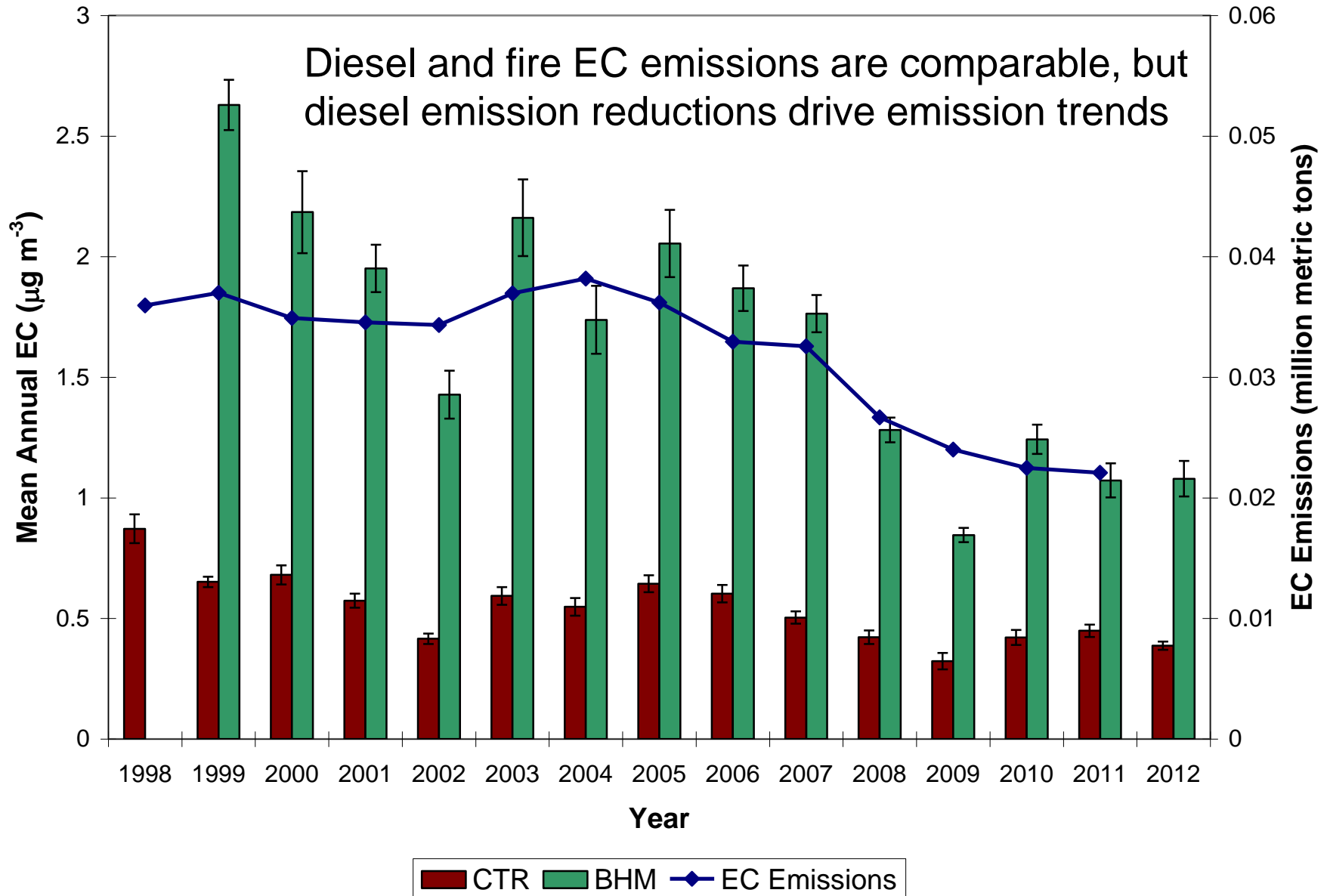
All SEARCH Sites Exhibit Declining Annual PM_{2.5} SO₄, EC, and OC – Why?



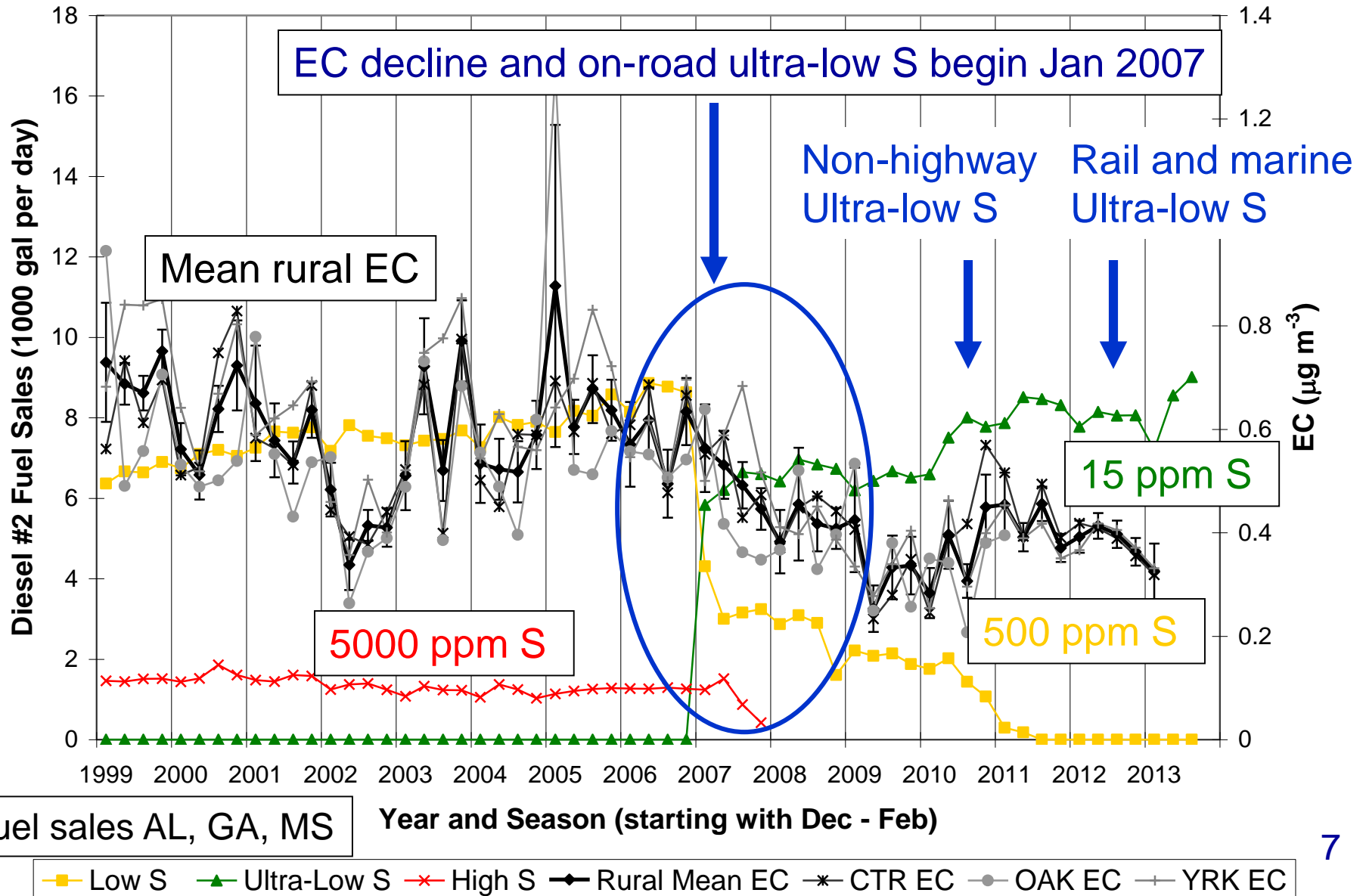
Gases (SO₂, NO_y, NO_z, NMOC, CO) and PM_{2.5} SO₄ Track Emissions



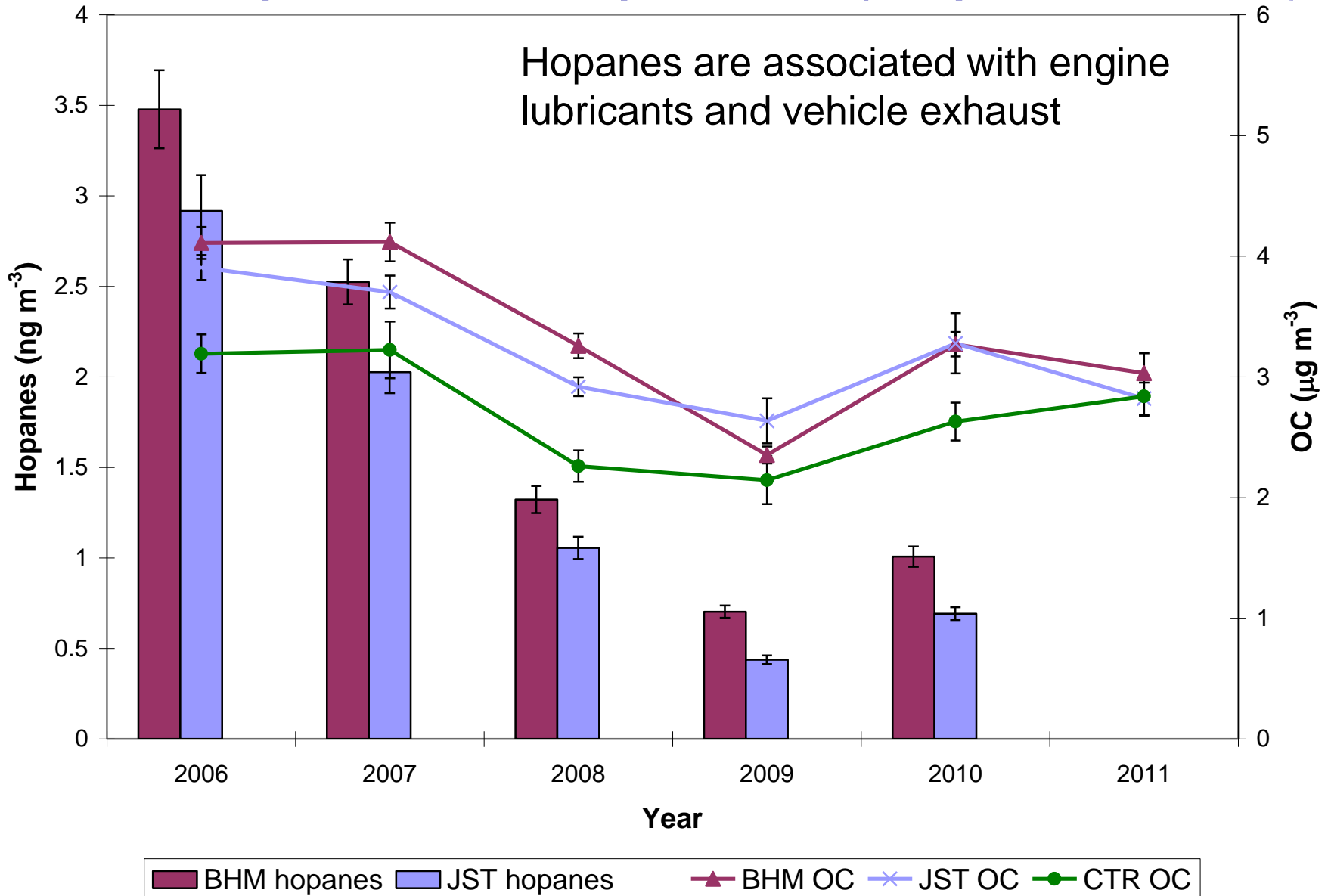
Are SEARCH EC Trends Consistent With EC Emissions? – Yes!



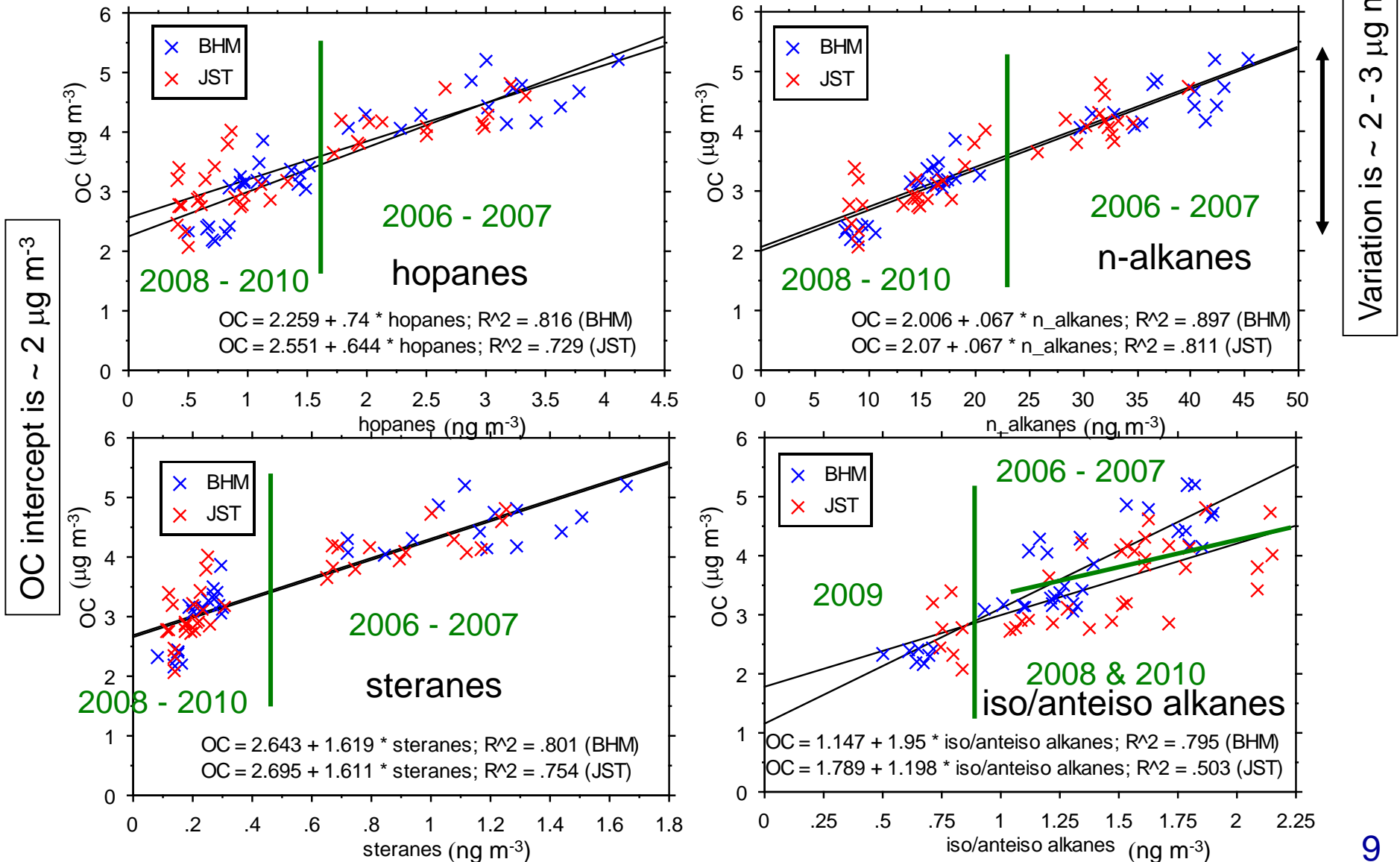
SEARCH Rural EC Decline Coincides With Introduction of Ultra-Low S Diesel Fuel



OC Trends Correlate With Decline in Non-polar OC Species (hopanes, etc)



Urban OC Varies with Non-polar Species (2 – 3 $\mu\text{g m}^{-3}$ Mobile PM OC + SOA?)



Data are 2006 – 2010 annual day-of-week means

Biomass Burning Event March 9 –10, 2014

Location

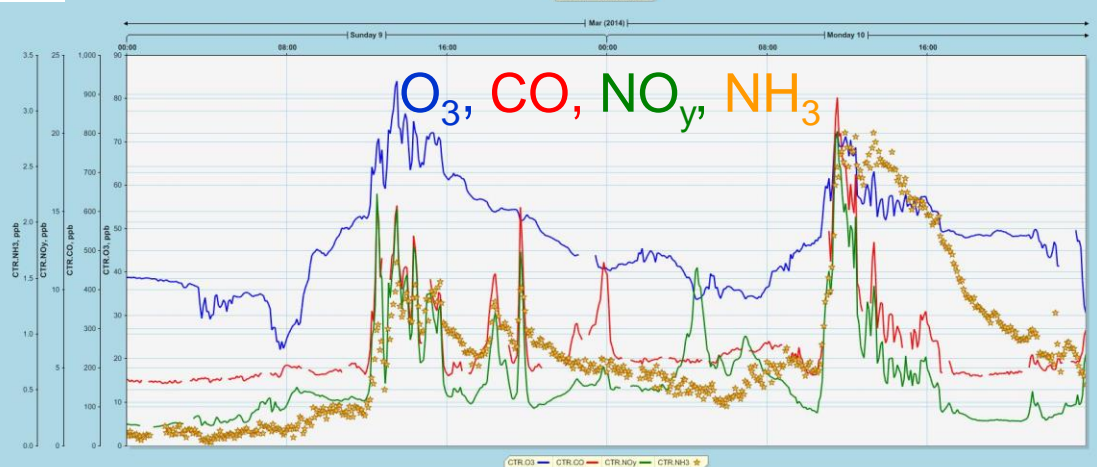
- ~10 km N of CTR
- ~ 1000 acres
- WDR ~ 300° (250 – 20)

EC and TC

- OC/EC 15:1 – 20:1 at peaks

Gases

- 8-hour O_3
 - 62 & 56 ppbv
 - 52 & 45 ppbv at BHM
- CO 300–900 ppbv



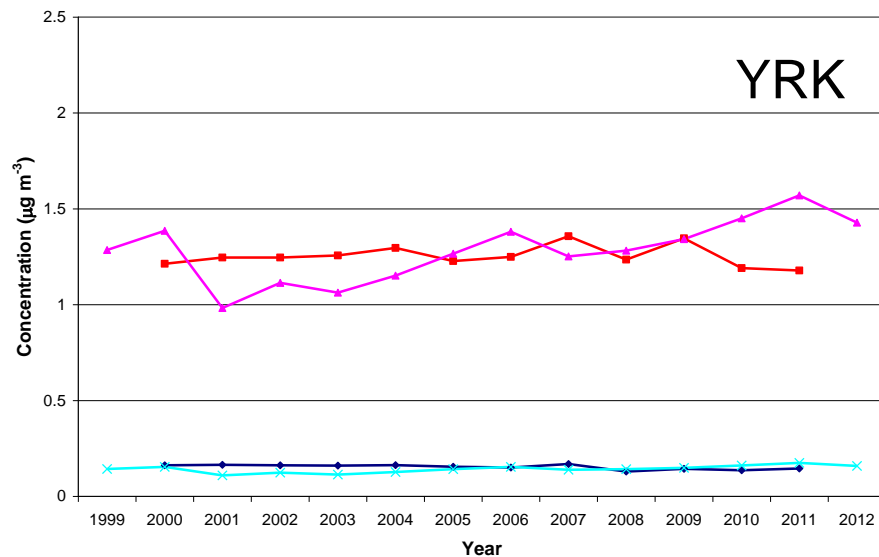
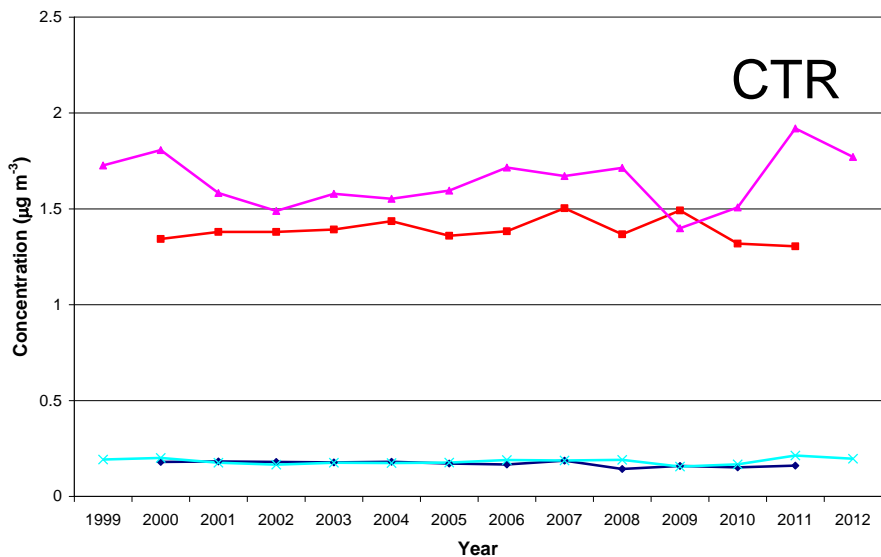
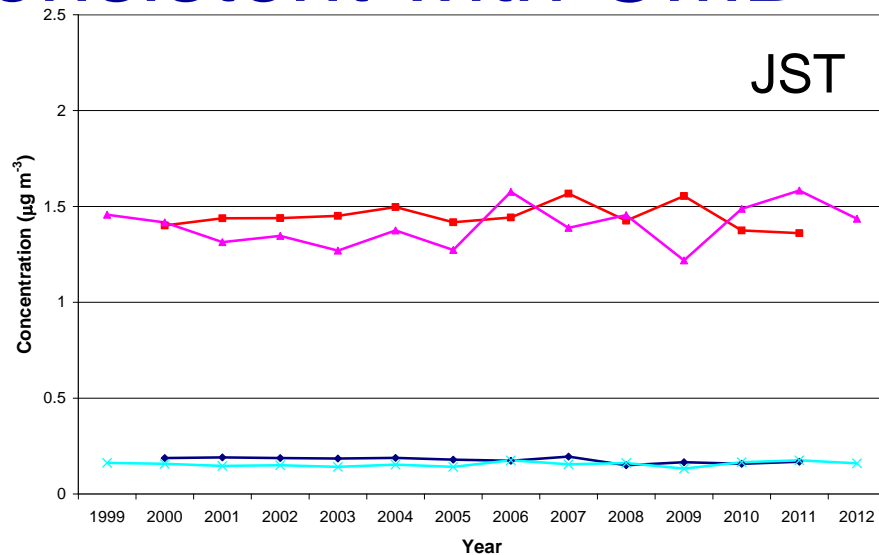
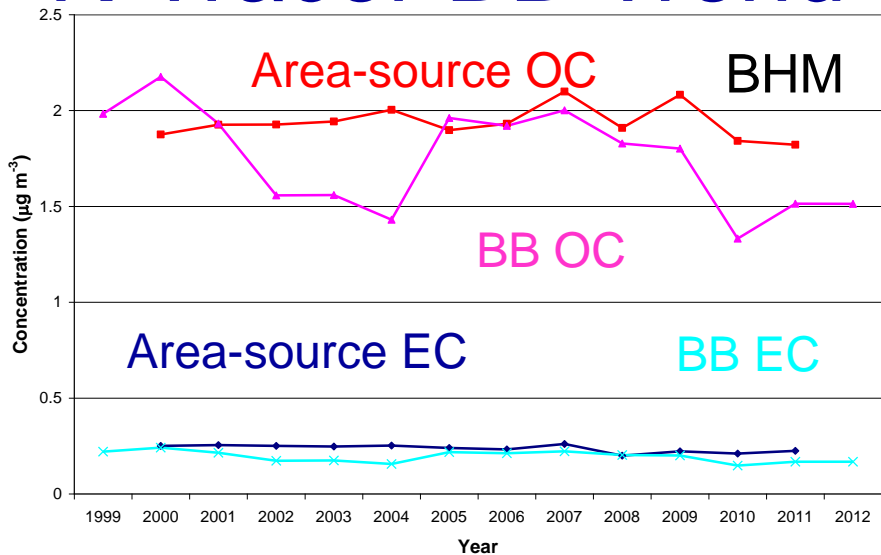
Application of Potassium Tracer Method to Estimate Biomass Burning OC

- Use K tracer approach*:
 - regress K against X** : $K = \alpha + \beta * X$
 - compute $K_b = K - \beta * X$
 - PM_{coarse} : tight $\Delta K / \Delta Si$ (0.10 – 0.13; $r^2 \sim 0.8$)
- $K_b > 0.4 \mu g m^{-3}$ are on 4th of July and Jan 1!
- Scale from K_b to BB TC, OC, and EC
 - TC/ K_b varies widely among fire types (wild \ll Rx)
 - Inventory annual average BB TC/ K_b 28:1 – 36:1
 - 2004-05 data: modern $\Delta TC / \Delta K_b \sim 22:1 - 82:1$

* Recently described by Pachon et al. 2013. *Atmos. Poll. Res.*

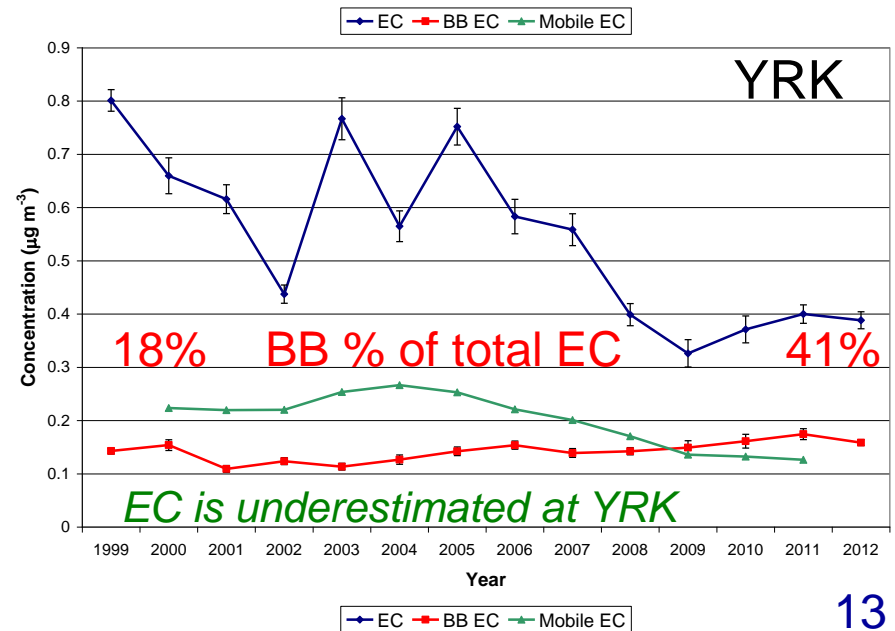
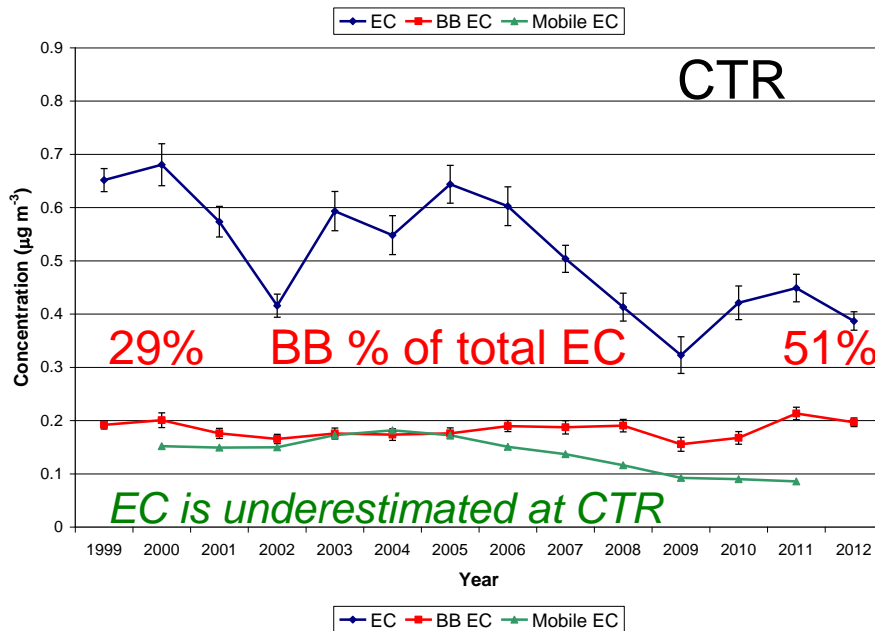
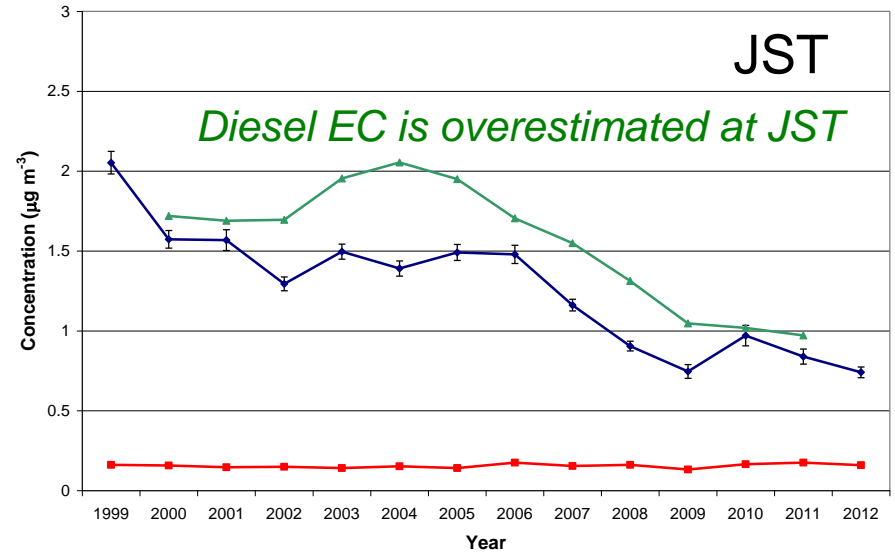
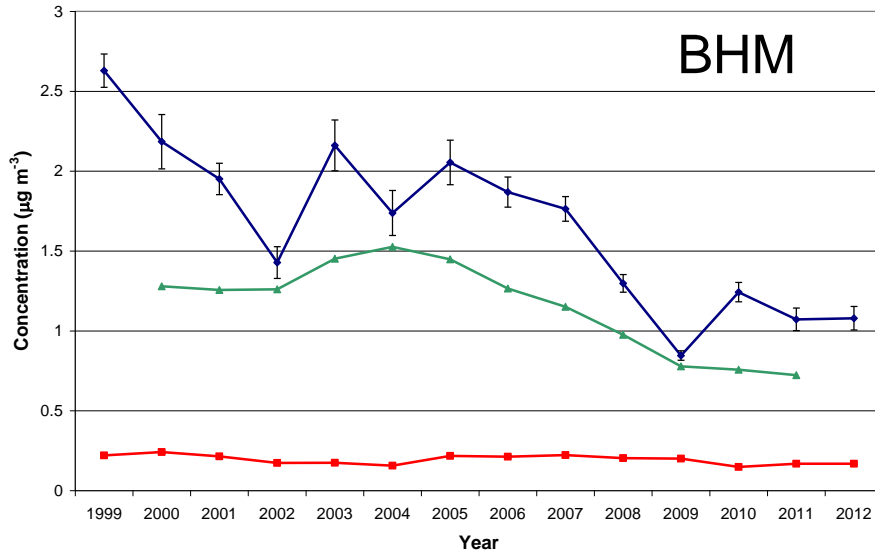
** X is an element that derives primarily from crustal material

K-Tracer BB Trend Consistent with CMB*



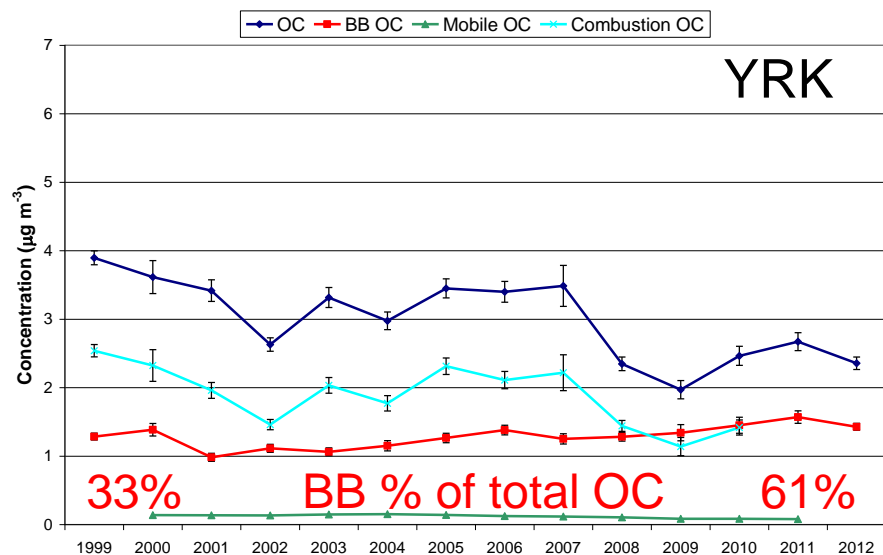
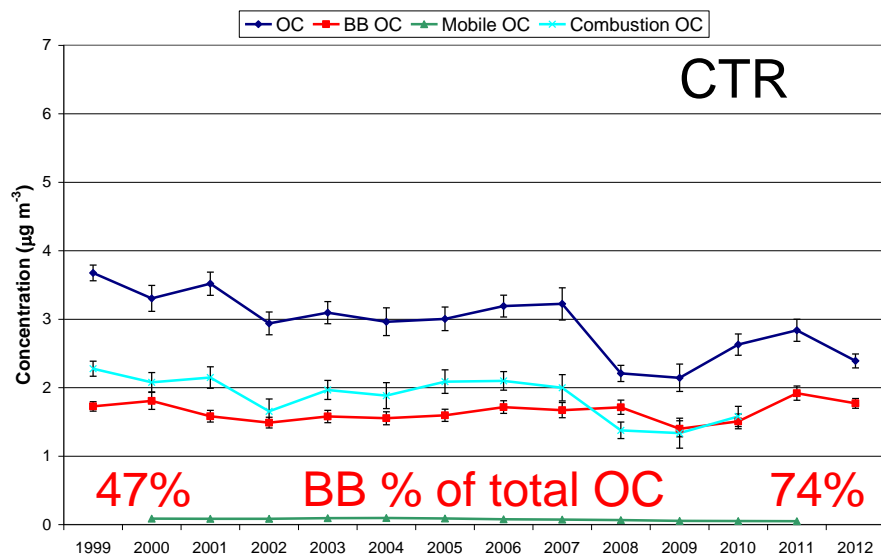
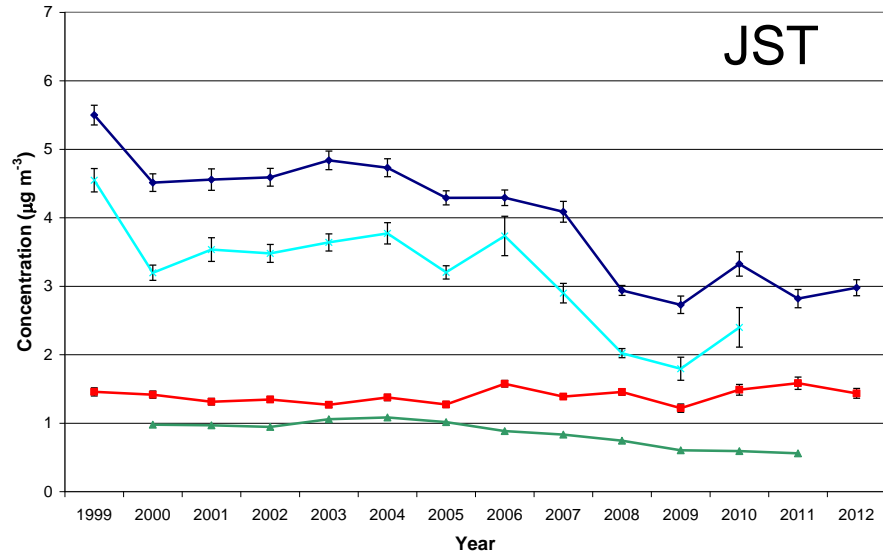
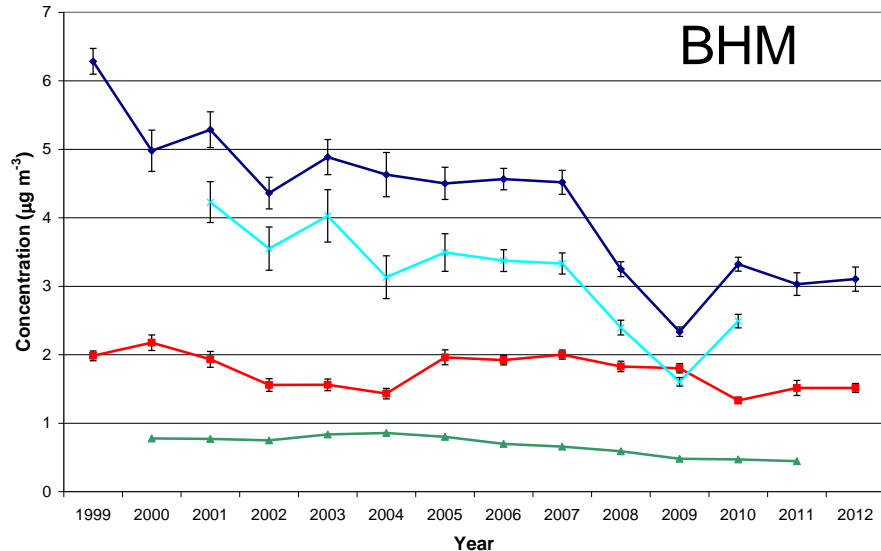
* Blanchard et al. 2013. *ES&T*. (Area source is predominantly fires)

BB EC ~ Constant but Increasing Fraction



Mobile EC (diesel + gasoline PM EC) from Blanchard et al. 2013. *ES&T*.

BB OC ~ Constant but Increasing Fraction



Mobile OC (diesel + gasoline PM OC) from Blanchard et al. 2013. *ES&T*.

Combustion OC is calculated from tracers EC, CO, non-soil K (Blanchard et al., 2008)

Summary:

Anthropogenic Influences on EC and OC

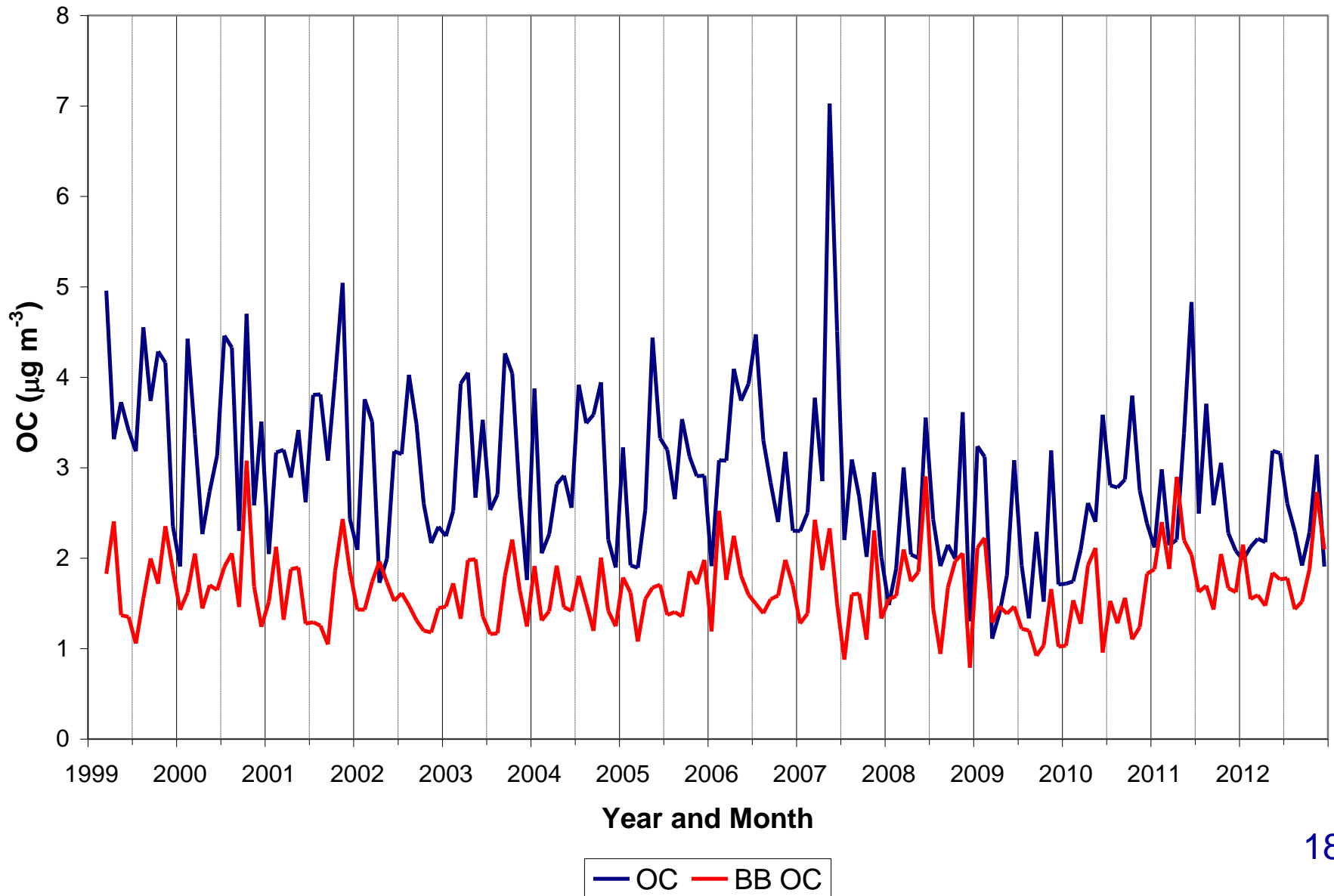
- Large reductions of anthropogenic emissions of SO_2 , NO_x , CO, VOC, EC, OC in SE US
- SEARCH ambient trends in SO_2 , NO_y , CO, NMOC, EC, OC, SO_4 , NO_3 track emission trends between 1999 (1992) and 2012
- SEARCH hourly and daily 1999 - 2012 measurements indicate that emissions from mobile sources and biomass combustion together are major contributors (~50 – 75%) to mean concentrations of EC and OC at CTR and at all SEARCH sites

SEARCH Presentations

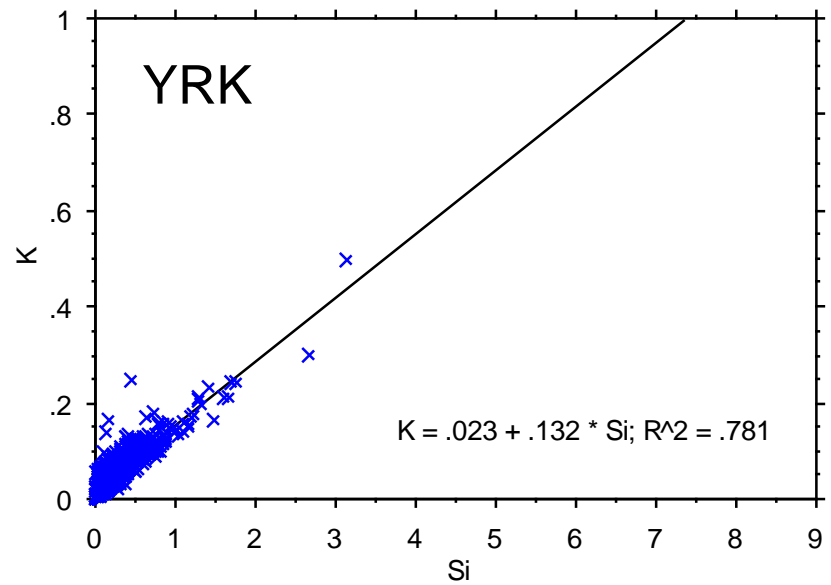
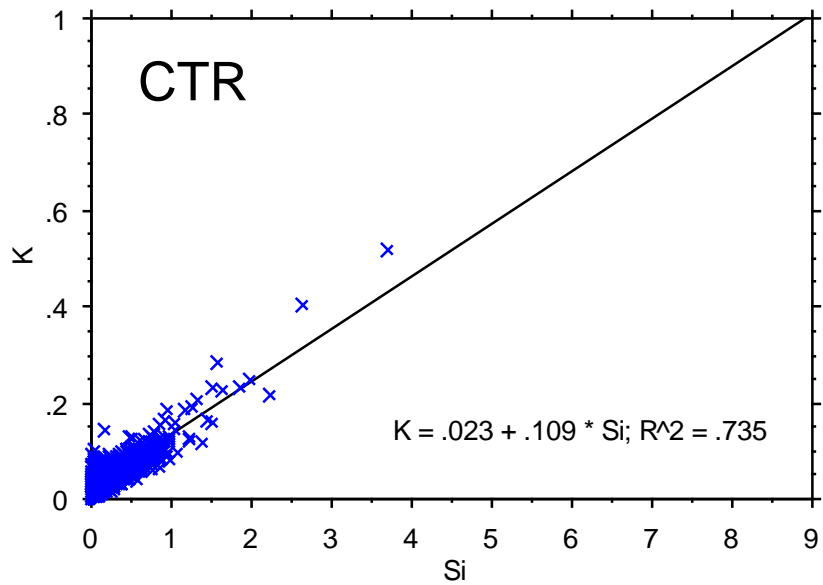
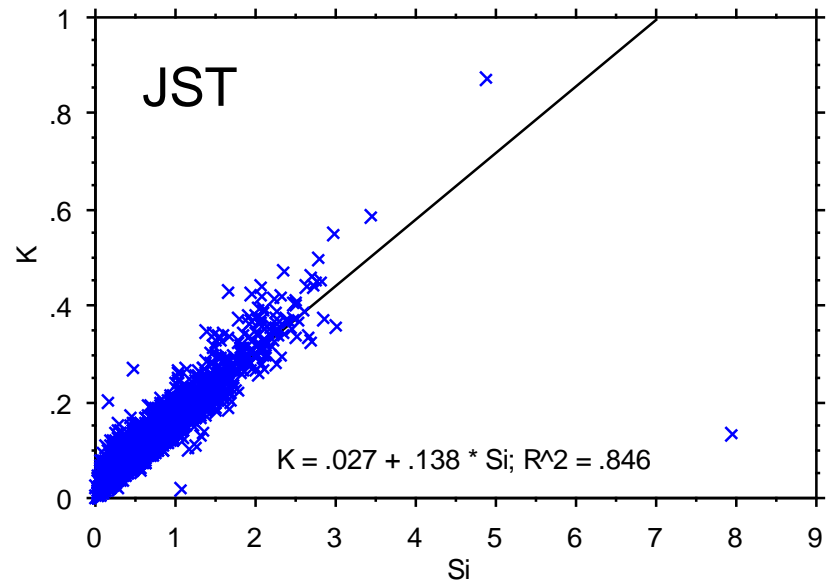
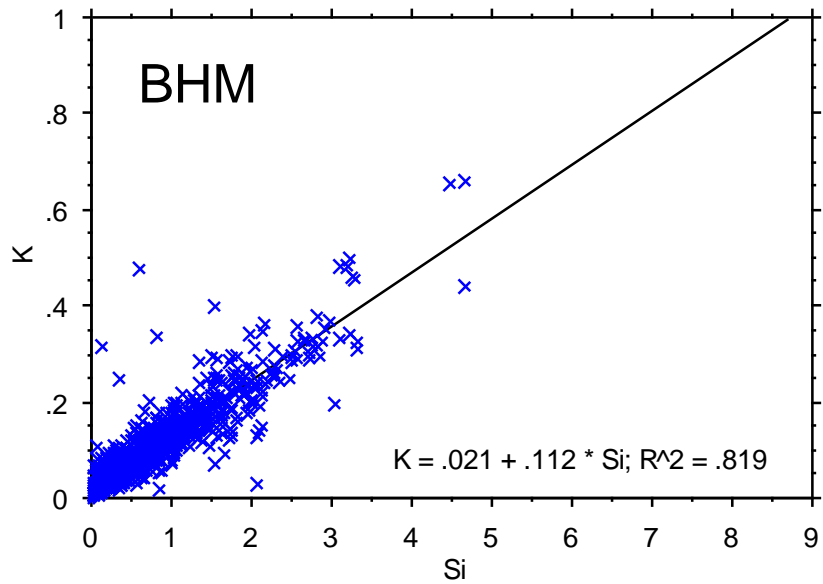
- Tuesday morning, Session 5
 - K. Baumann et al.: *An Historical Perspective on Air Quality at CTR in Jun-Jul*
 - S. Shaw: *Outstanding Questions on Organic Aerosols*
- Poster Session 2
 - E. Edgerton et al.: *First Look at ^{14}C Data During the Centreville, AL SOAS Campaign*
 - G. Hidy et al.: *Chemical Climatology of the Southeastern U.S., 1999 - 2013*

Extras

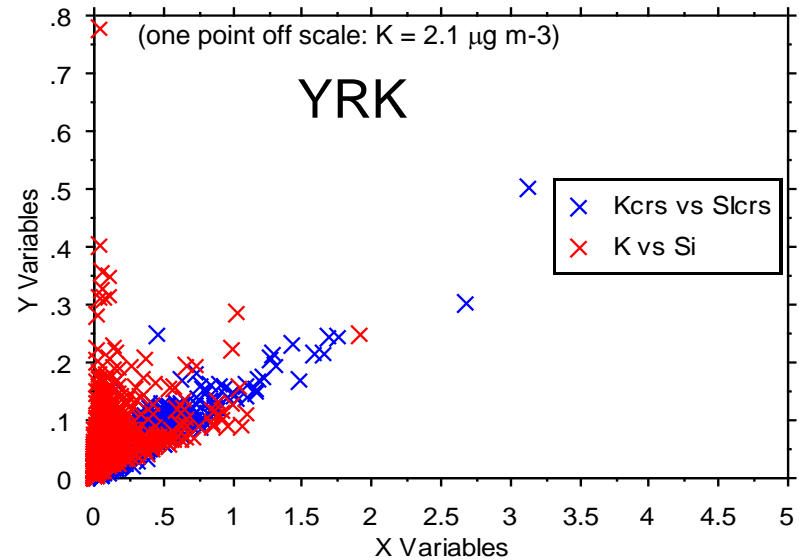
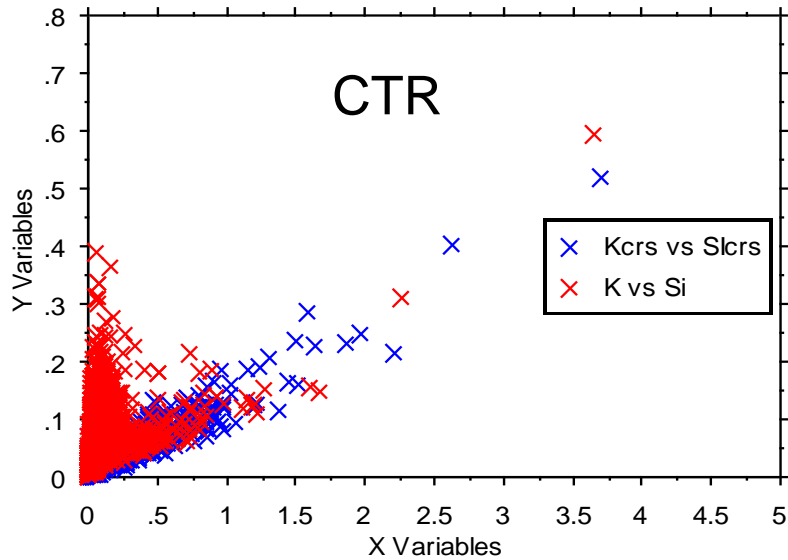
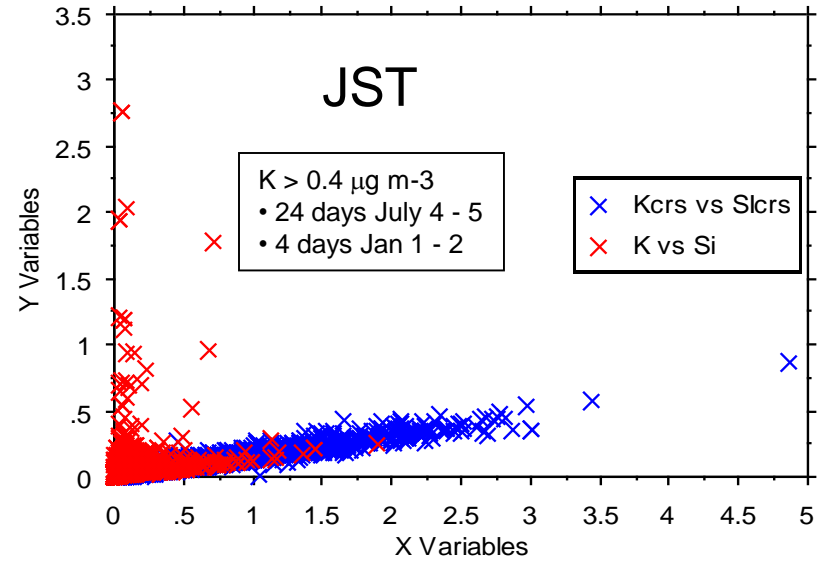
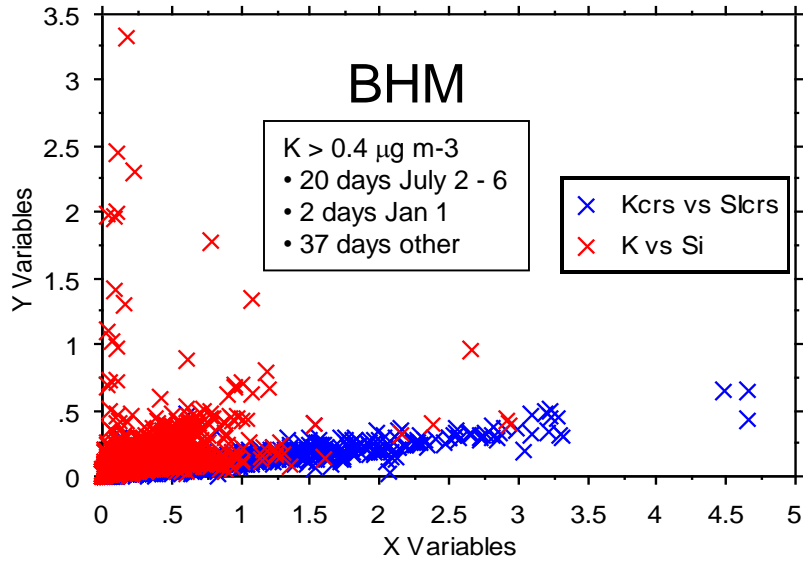
CTR Monthly Average OC and BB OC



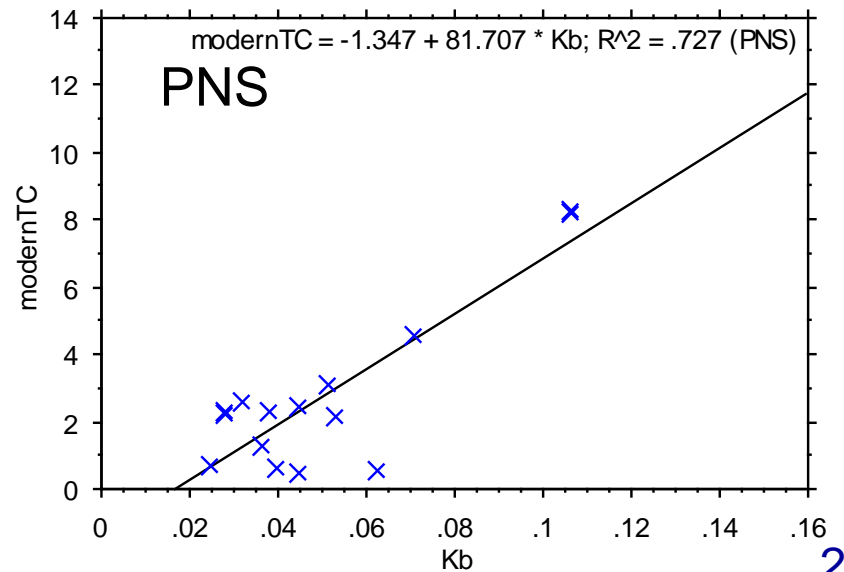
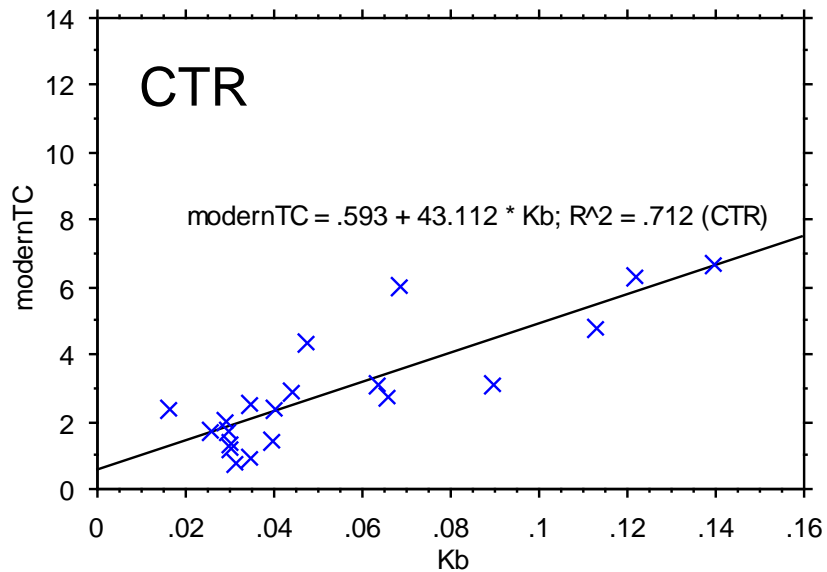
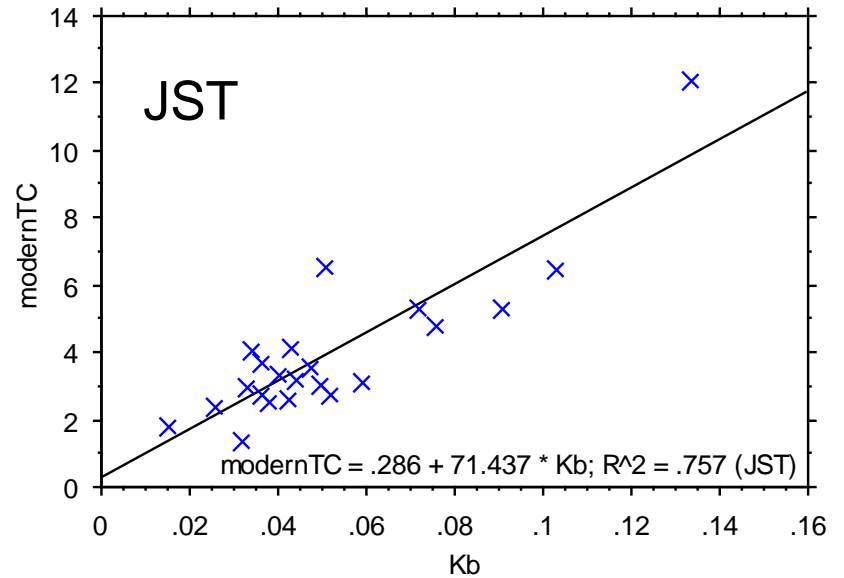
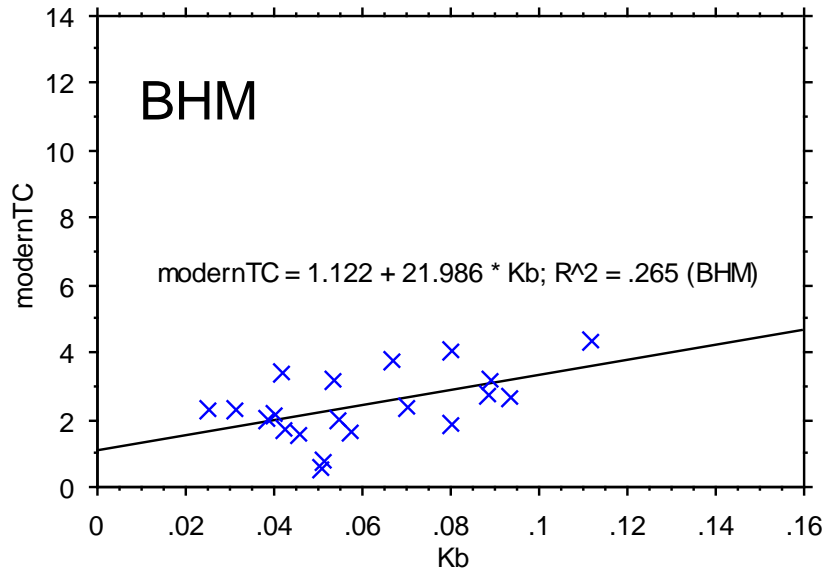
Correlation between K and Si in PM_{coarse}



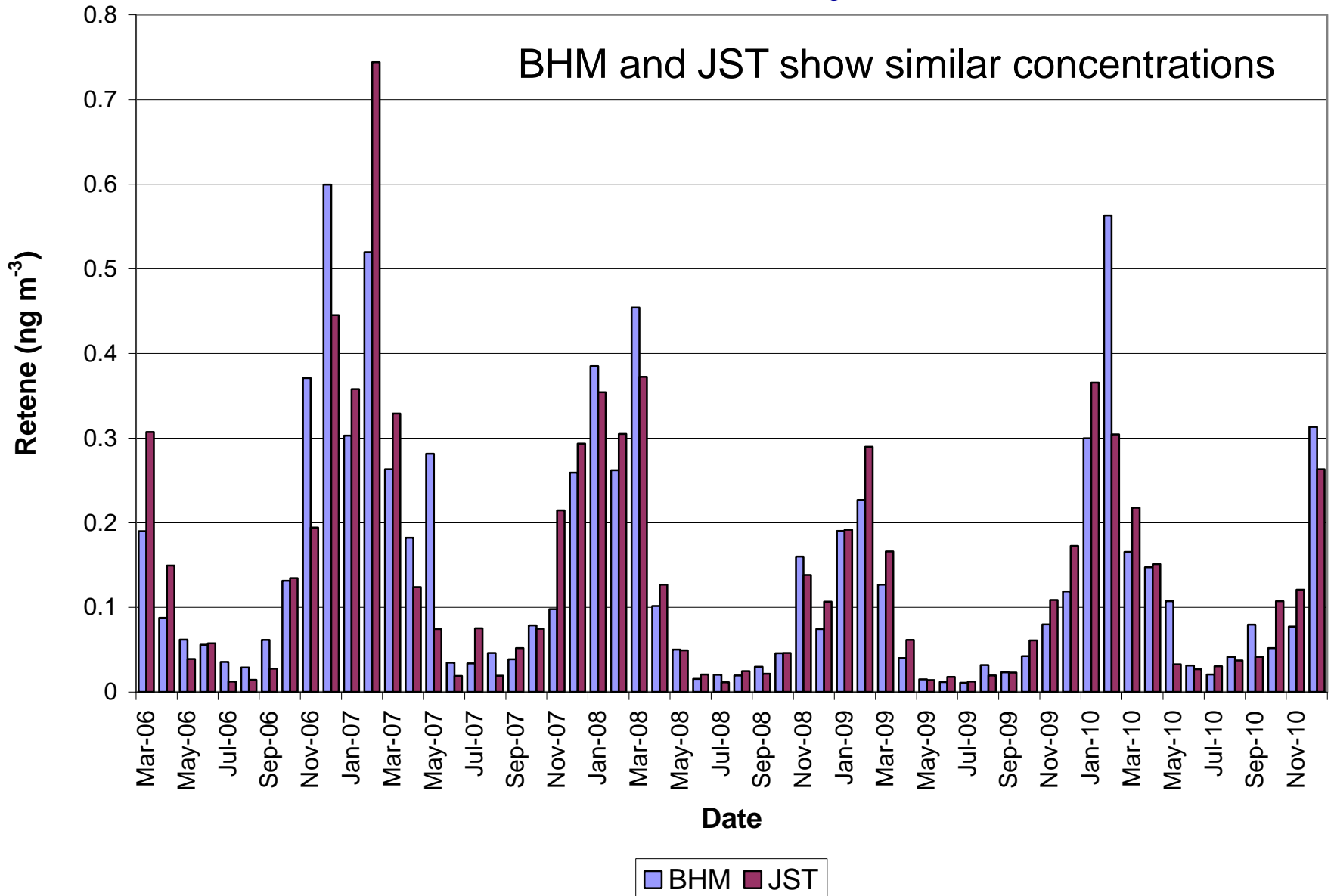
Non-soil K Sources in PM_{2.5}



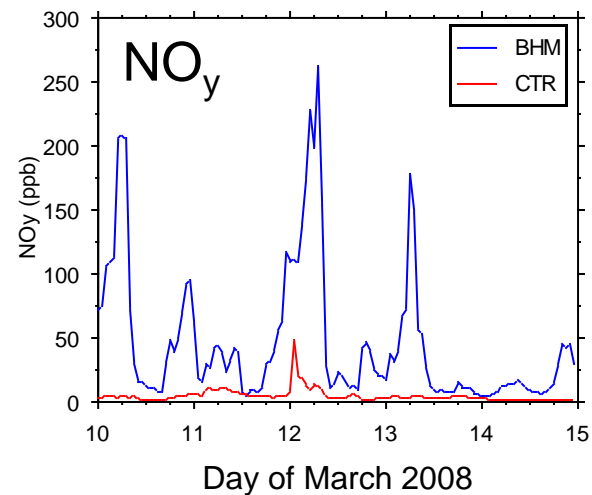
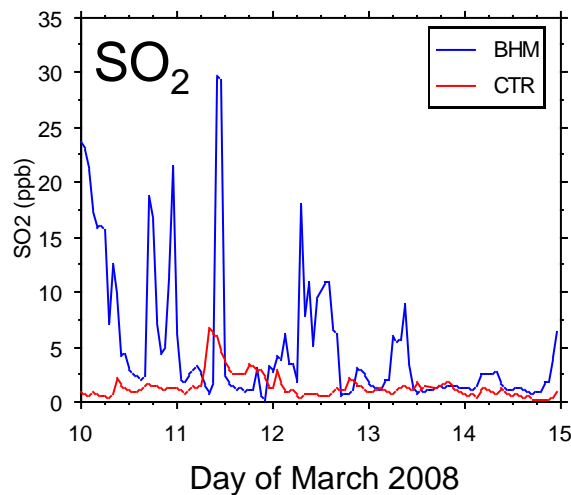
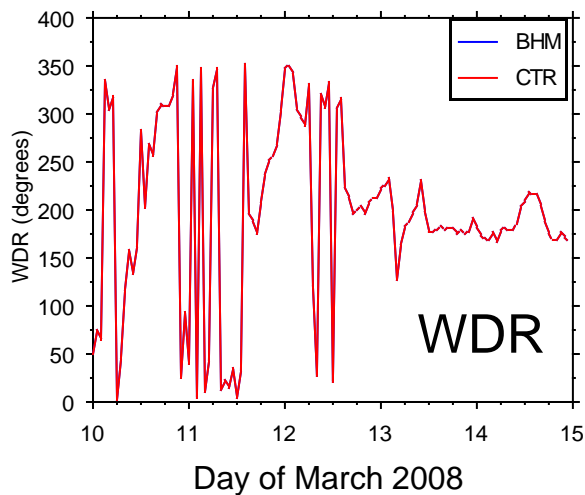
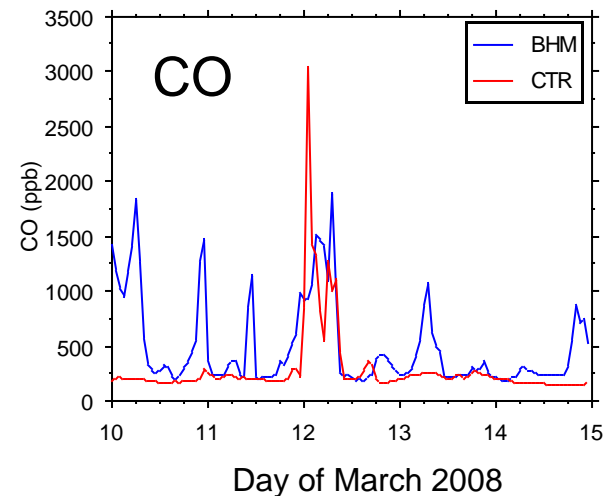
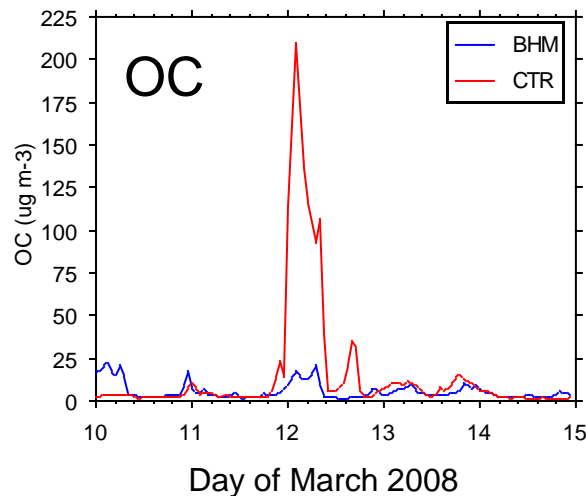
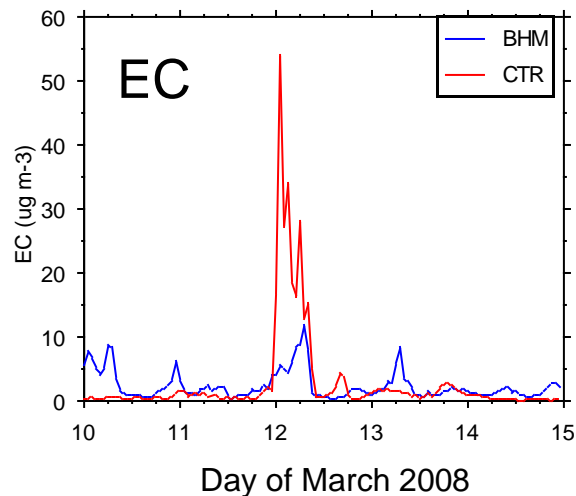
Kb vs. ModernTC by Site (Oct – Apr)



Tracers of Biomass Burning are Seasonal but Always Present

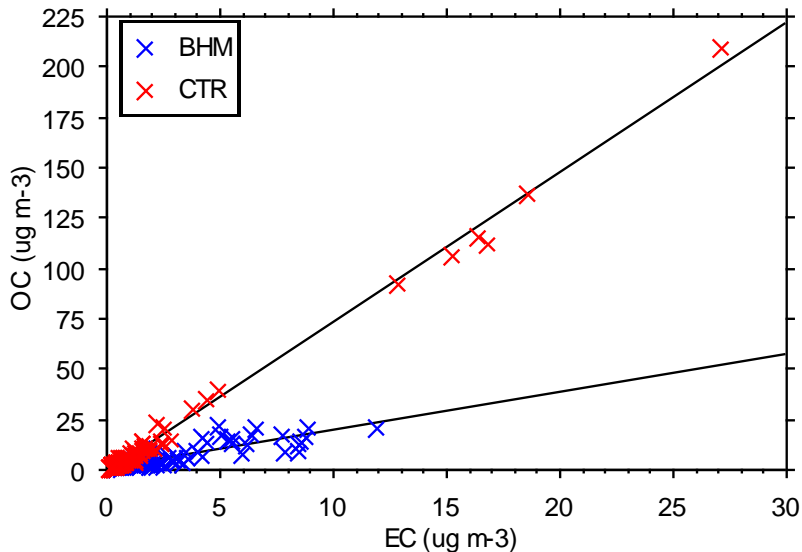


Example Biomass Burning Event March 11 – 12, 2008

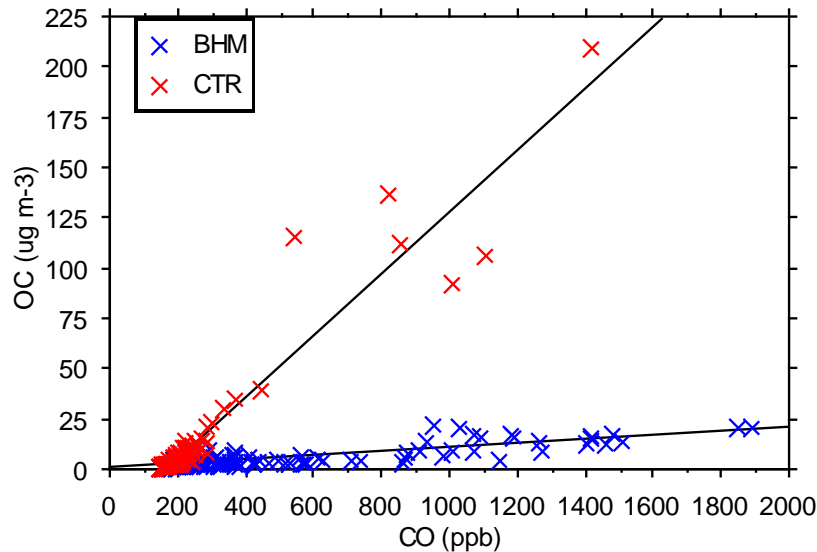


EC, OC, CO enhanced at CTR – some excess NO_y – no excess SO₂

Characteristics of the 3/12/2008 Event

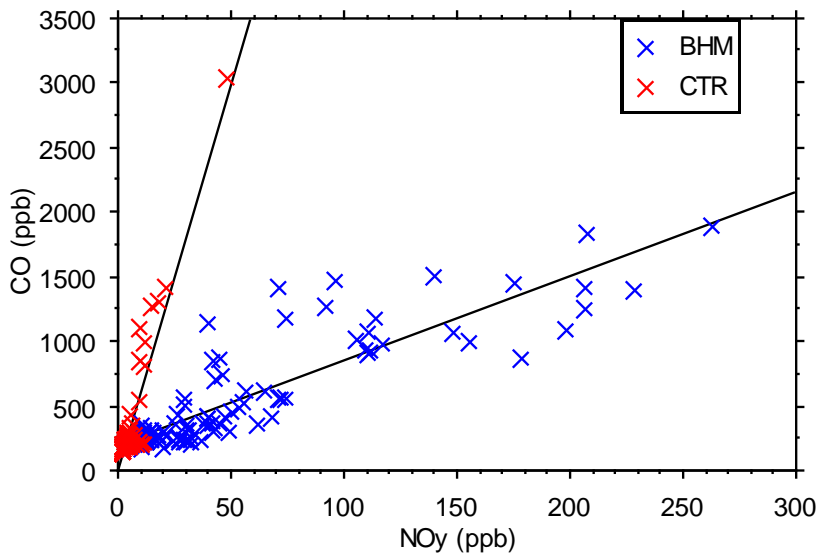


$OC (ug m^{-3}) = 1.361 + 1.881 * EC (ug m^{-3}); R^2 = .72$ (BHM)
 $OC (ug m^{-3}) = -.862 + 7.395 * EC (ug m^{-3}); R^2 = .994$ (CTR)



$OC (ug m^{-3}) = .769 + .01 * CO (ppb); R^2 = .692$ (BHM)
 $OC (ug m^{-3}) = -25.261 + .153 * CO (ppb); R^2 = .912$ (CTR)

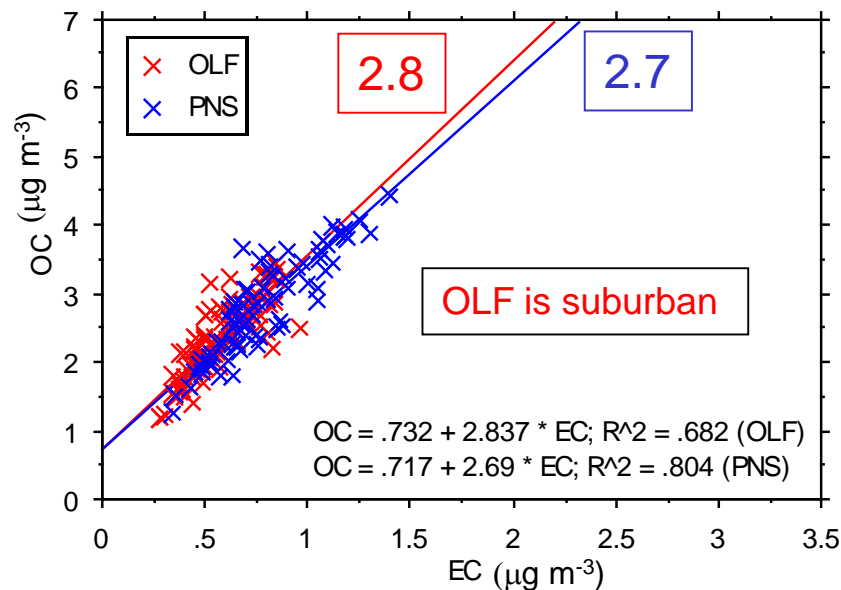
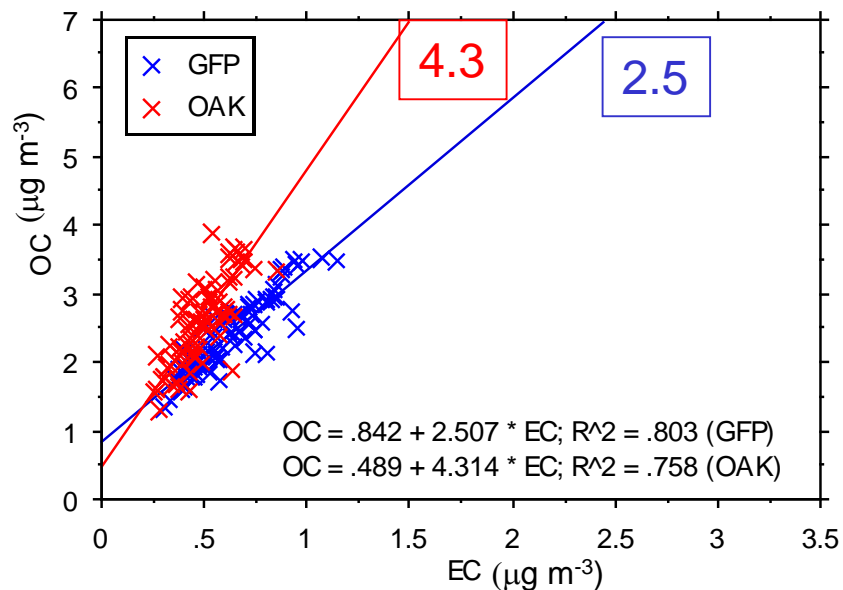
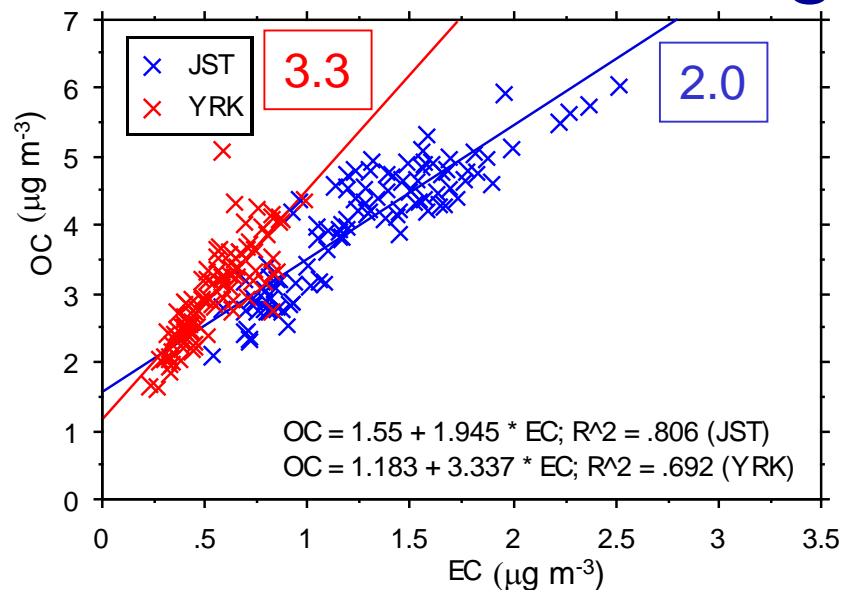
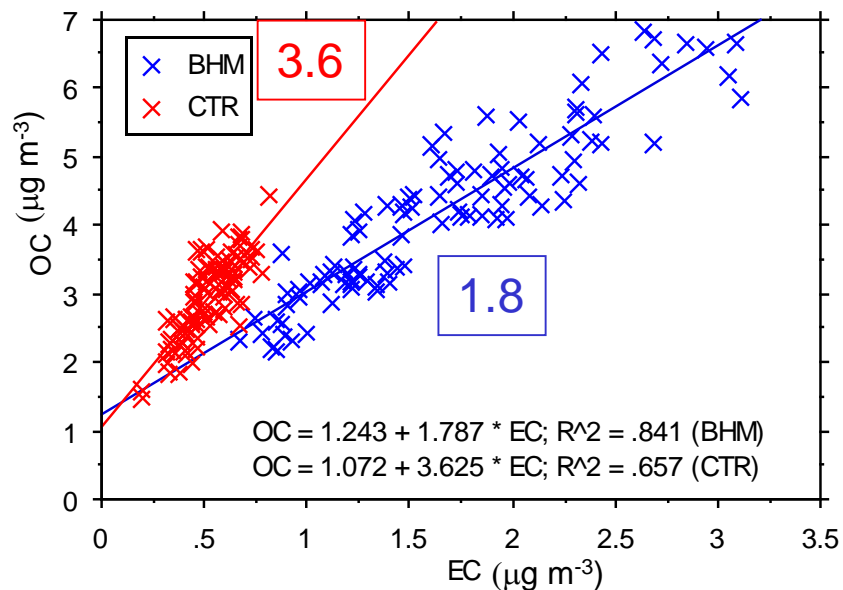
- CTR $\Delta OC / \Delta EC = 7.4$
- BHM $\Delta OC / \Delta EC = 1.9$
- CTR $\Delta CO / \Delta NO_y = 60.0$
- BHM $\Delta CO / \Delta NO_y = 6.5$



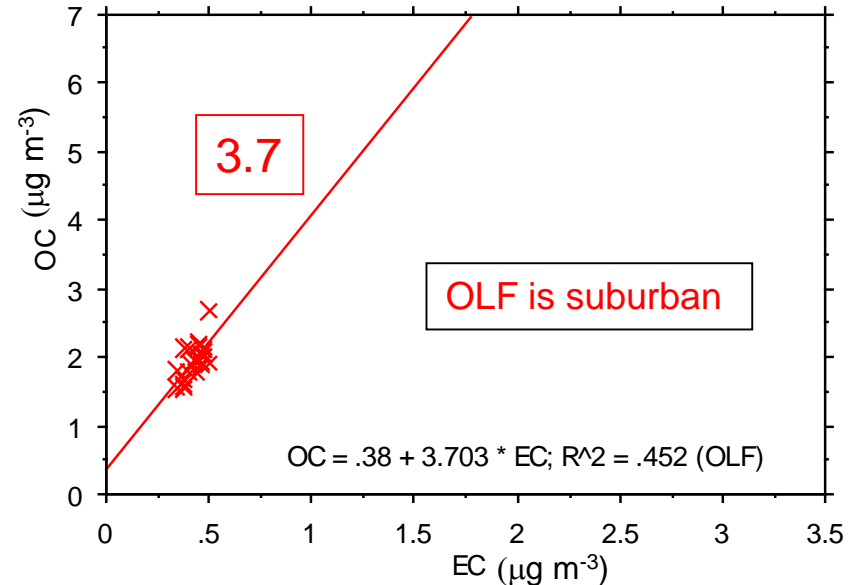
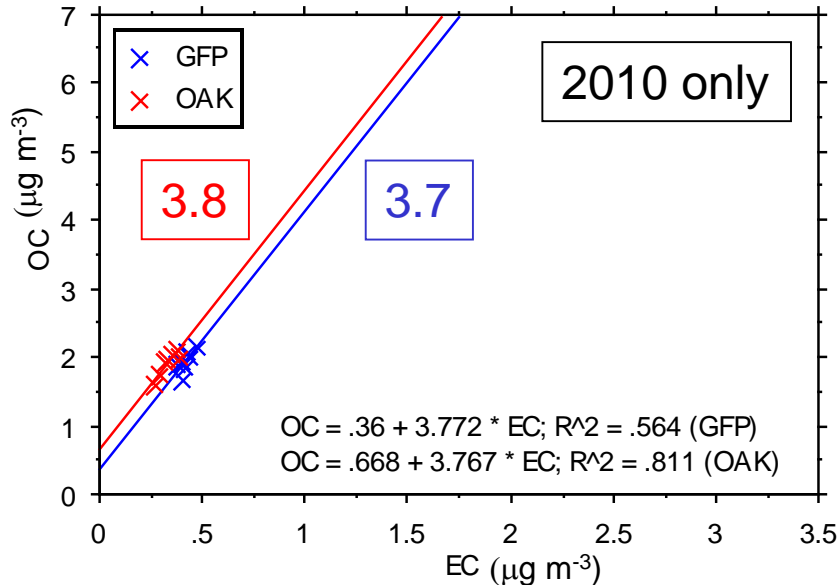
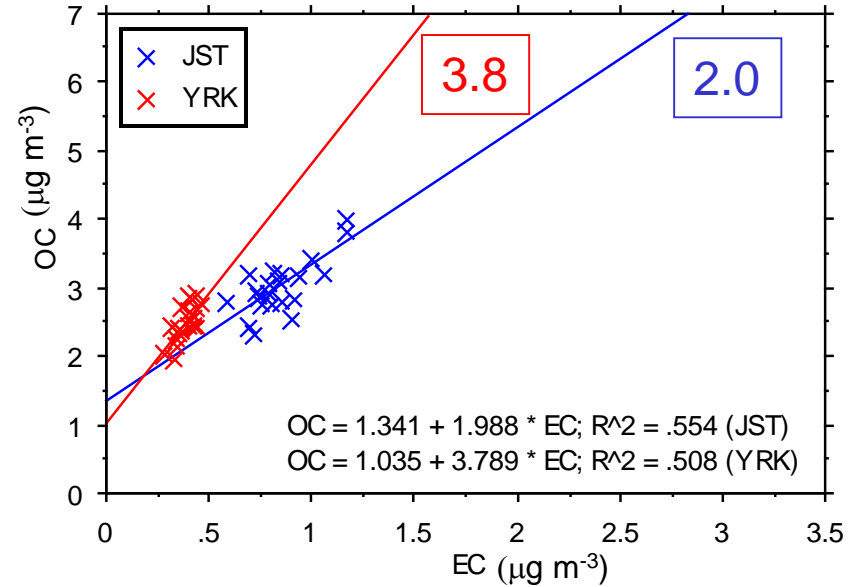
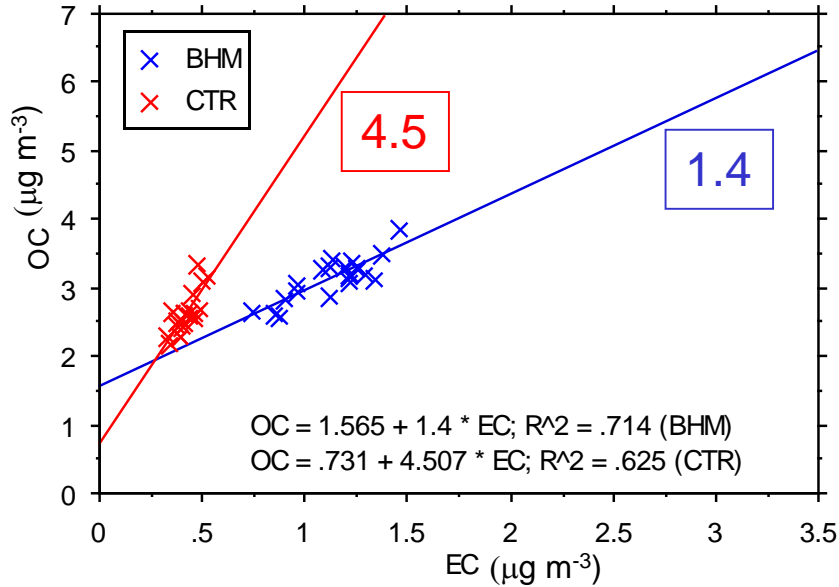
$CO (ppb) = 195.171 + 6.493 * NO_y (ppb); R^2 = .791$ (BHM)
 $CO (ppb) = -10.438 + 59.954 * NO_y (ppb); R^2 = .827$ (CTR)

1999 – 2012 Δ OC/ Δ EC Signatures: Urban Mobile & Rural Biomass Burning

OC intercept is $\sim 1 \mu\text{g m}^{-3}$

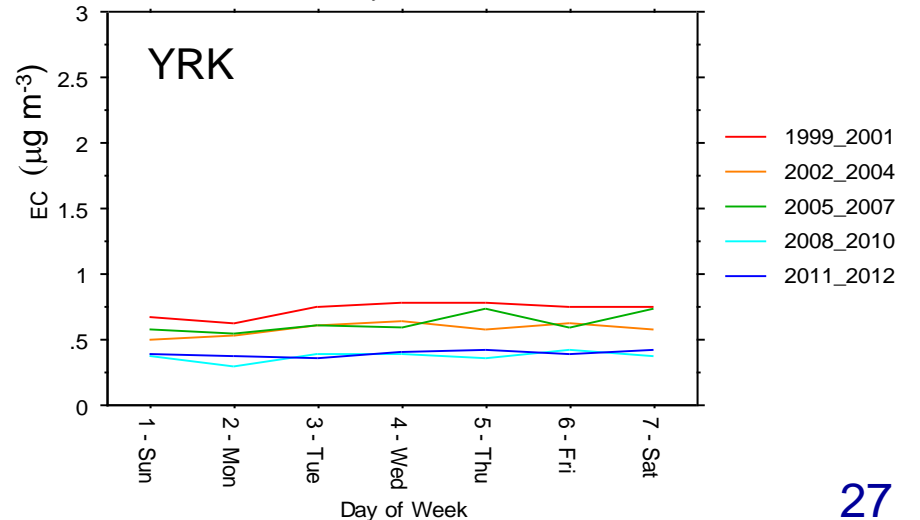
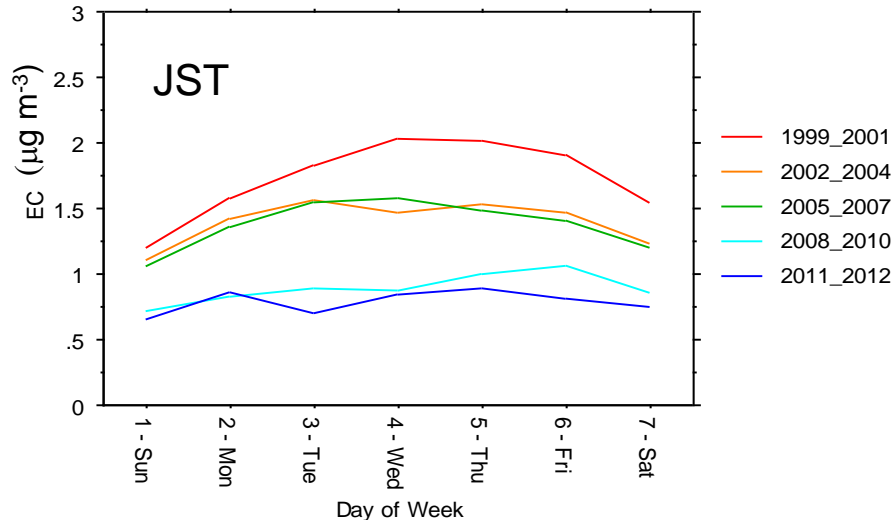
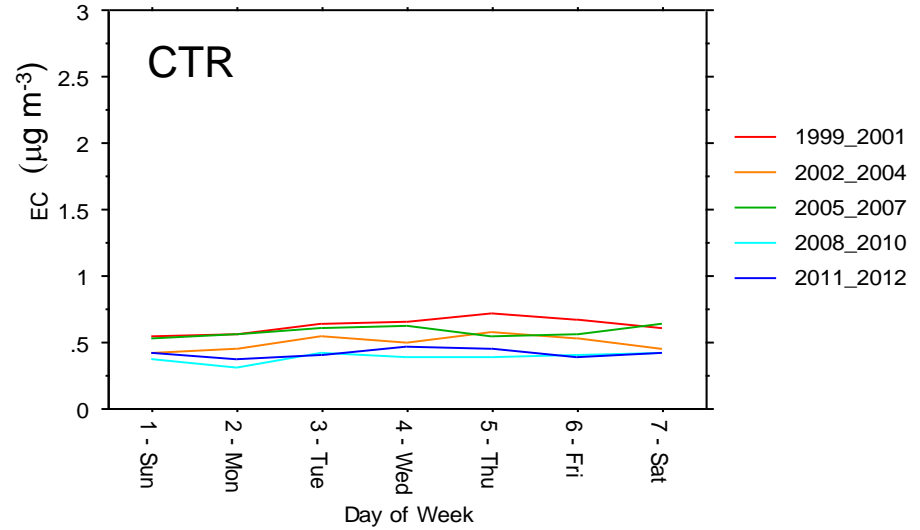
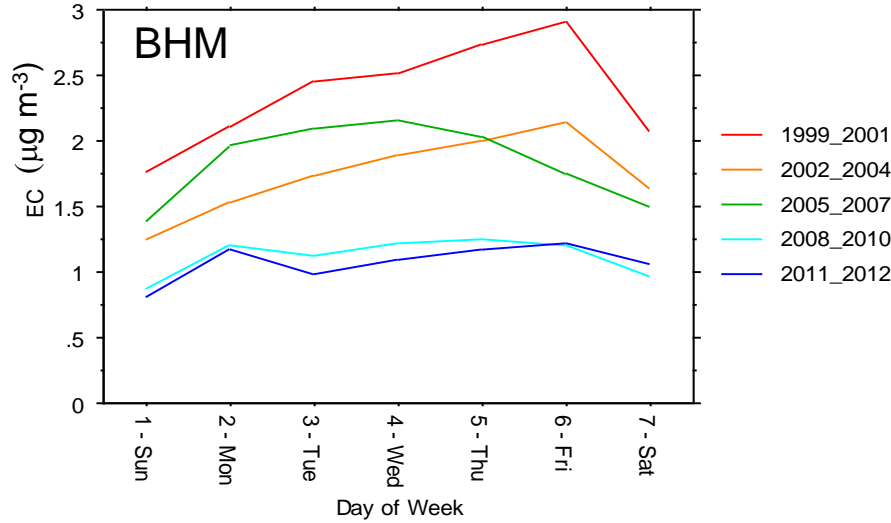


2010 – 2012: Reduced EC and OC with More Similar Urban & Rural OC Range



Urban Excess EC and Day-of-Week Variations Declined from 1999 to 2012*

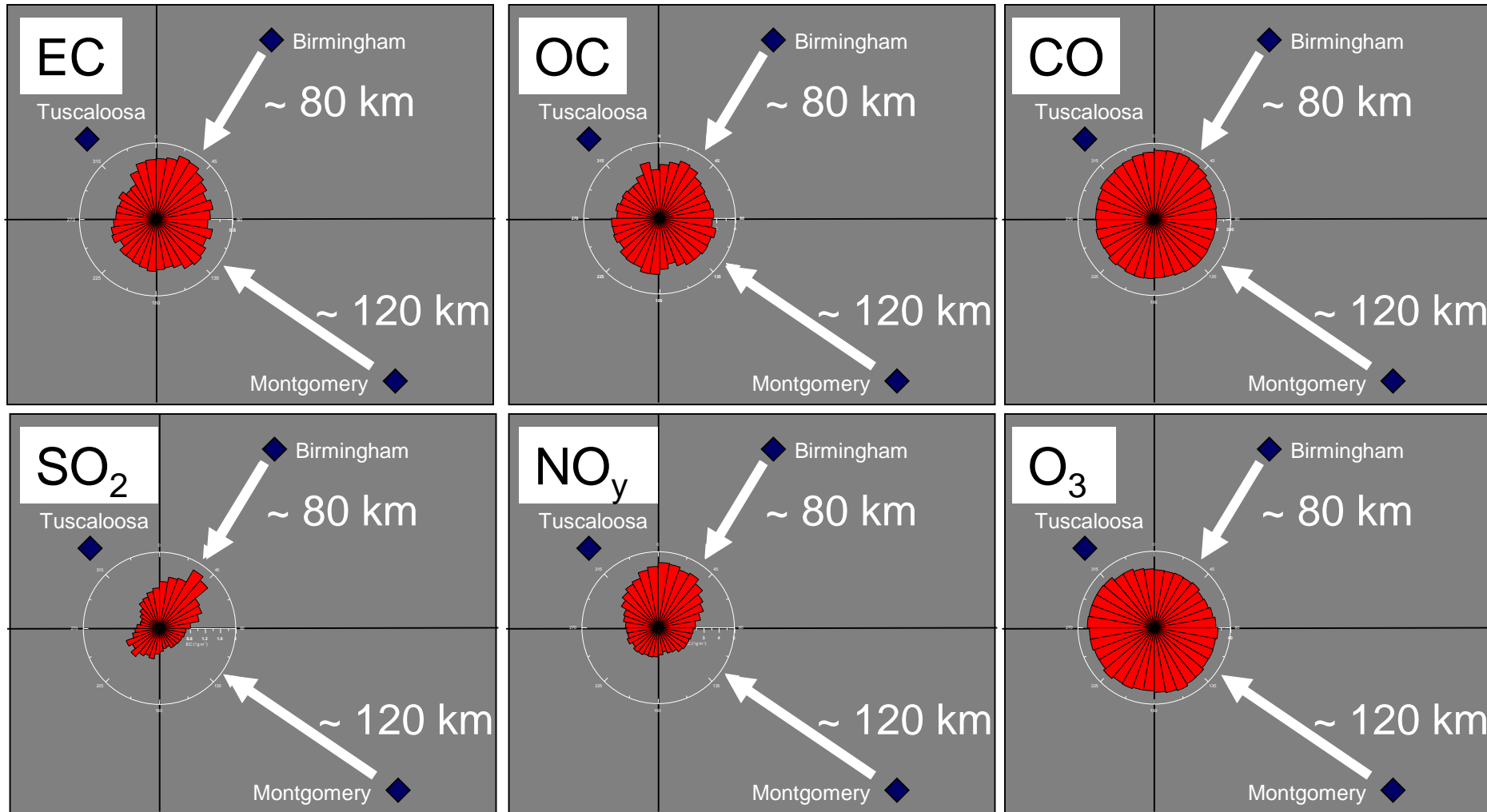
Lower EC post-2007 reflects diesel PM emission reductions



*DoW EC Variations are Linked to HD Diesel Traffic Patterns

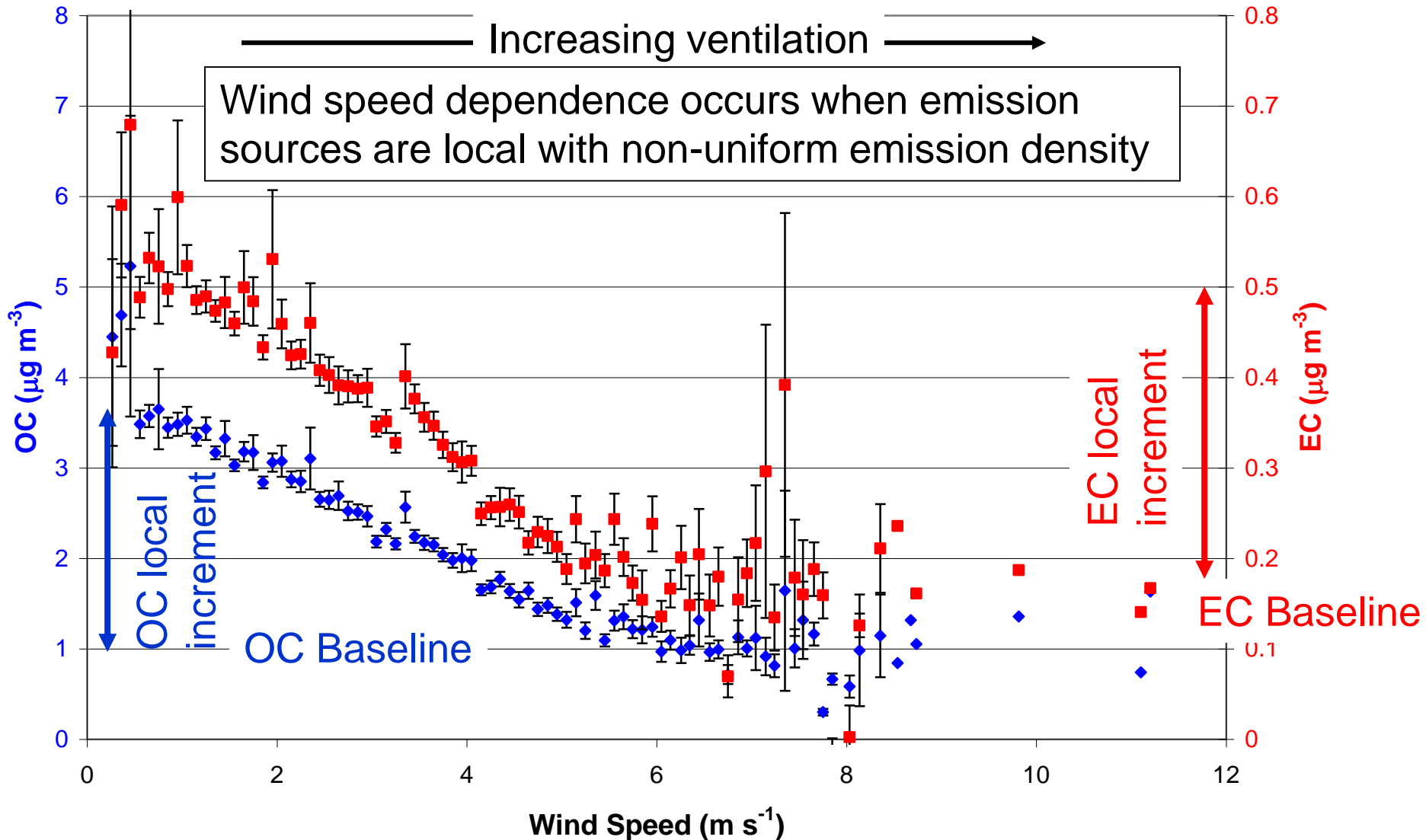
CTR EC, OC, CO, SO₂, NO_y Higher With Winds From Birmingham & Montgomery*

Directionality implies local source contributions



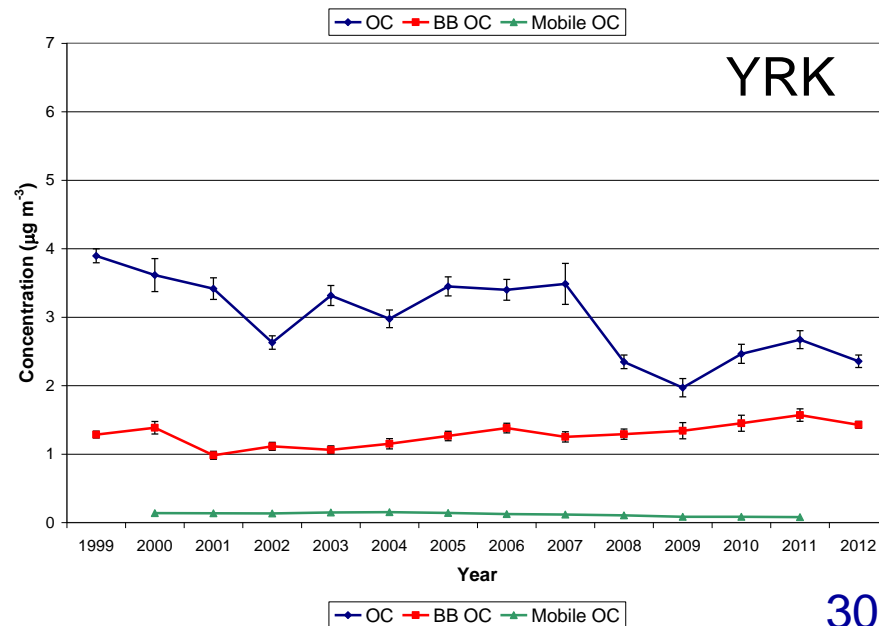
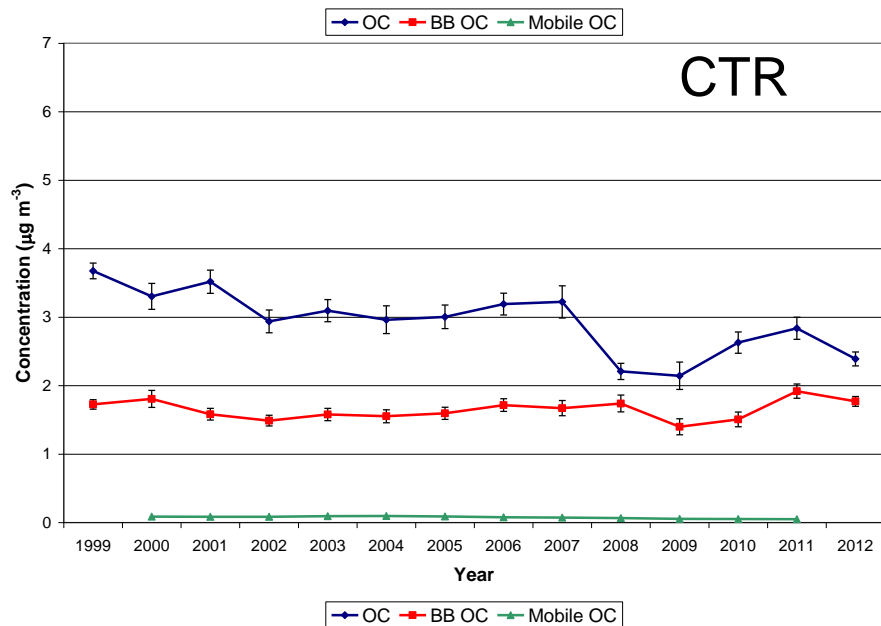
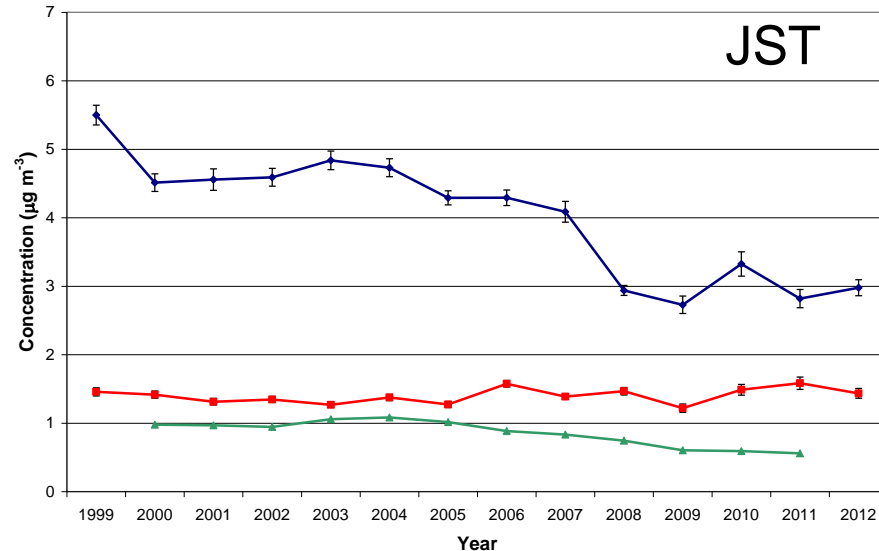
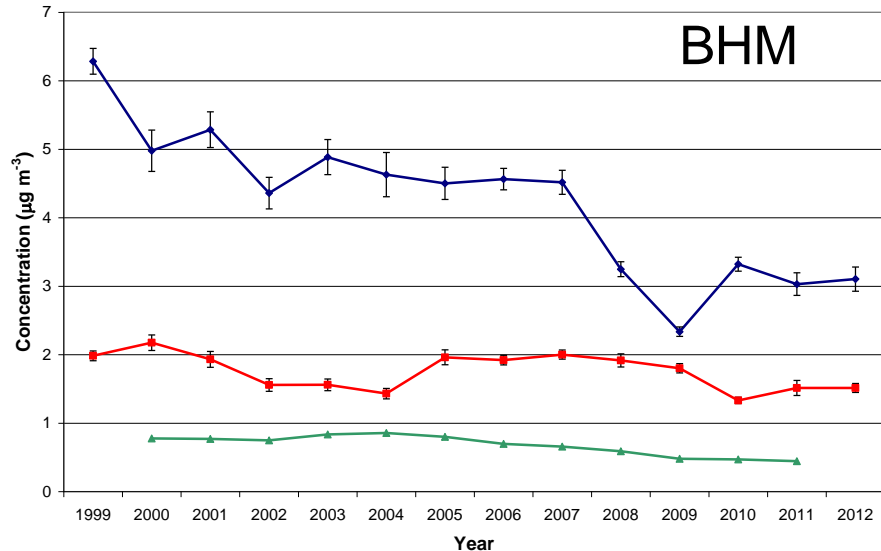
* All hourly data, 2008 - 2010

Mean CTR Hourly EC & OC Decline with WS to Baseline Levels



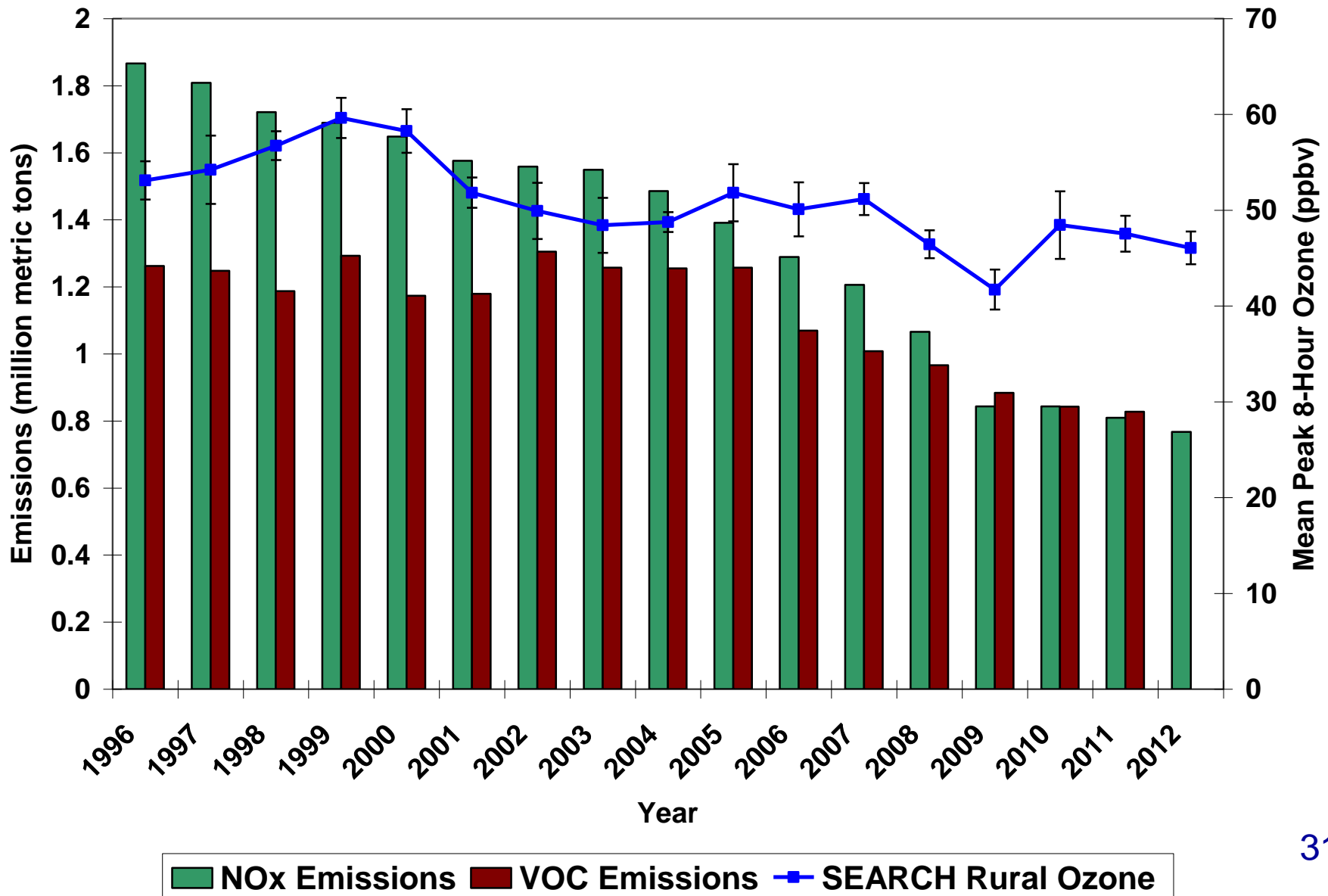
2008 – 10 data in 0.1 m s^{-1} bins

BB OC ~ Constant but Increasing Fraction



Mobile OC from Blanchard et al. 2013. *ES&T*. (diesel + gasoline PM)

What Hasn't Changed (Much)? – O₃!



Mean Concentrations n-alkanes

