Emissions and Evolution of Trace Gases and Particles in Agricultural Fire Plumes

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1 Introduction

- Agricultural fire emissions can have a large impact on atmospheric composition and air quality on regional scales.
- Chemical and physical transformations of primary emissions can lead to significant changes in gaseous and particulate phase compositions of the smoke.
- Emissions and smoke chemistry are not well characterized.

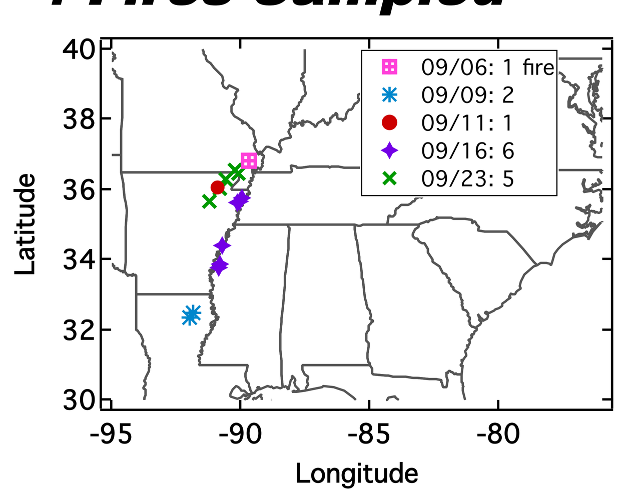
2 Objectives

- To quantify emissions of trace gases and fine particles from 15 agricultural fires.
- To study the evolution of NO_y species (PAN, NO_x, HNO_3, nitrate), O_3, organic aerosol (OA), and brown carbon (BrC) in fire plumes.

3 Aircraft Instrumentation

Gas	SO_2, HCl, PAN	GaTech CIMS
	HCN, Hydroxyacetone, $C_5O_3H_8$, HNO_3	Caltech CIMS
	VOC and OVOC	Innsbruck PTR-MS
	NO_x, NO_y, O_3	NOAA Chemiluminescence
	CO_2	NASA AVOCET
	CO	NASA DACOM
	CH_2O	Laser-induced fluorescence
Aerosol	BC	NOAA SP2
	SO_4, NO_3, NH_4, Cl, OA	CU HR-ToF-AMS
Optical	Particle absorption coefficients	NASA PSAP

4 Fires Sampled



Vegetation: rice straw

5 Analysis Methods

- Normalized excess mixing ratio (NEMR) was used to calculate emission factors and to study evolution

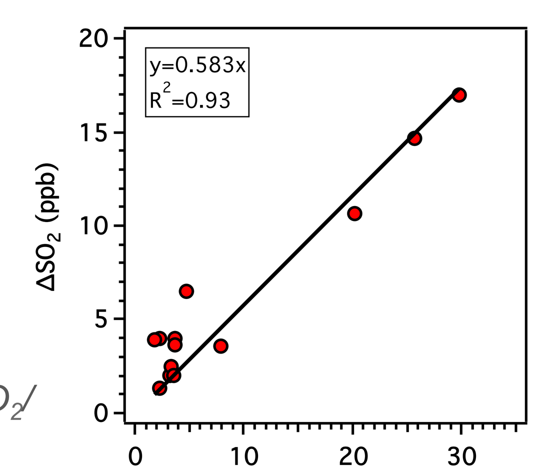
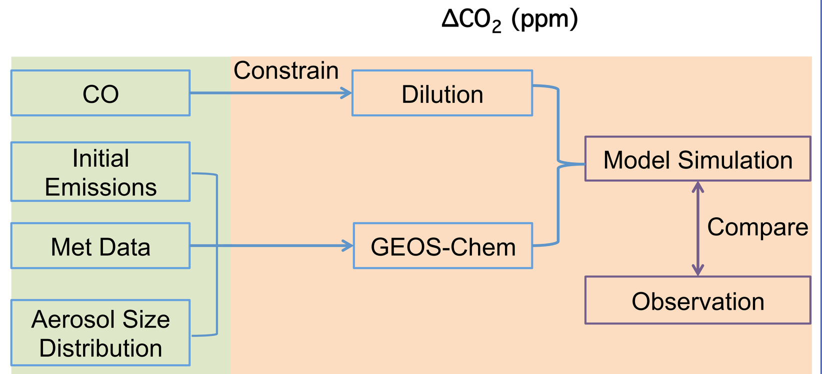


Figure 1. Emission ratio plot of ΔSO_2/ ΔCO_2 from fire #1 on Sept 11.

- A Lagrangian box model was used for modeling smoke chemistry



5 Emission Factors

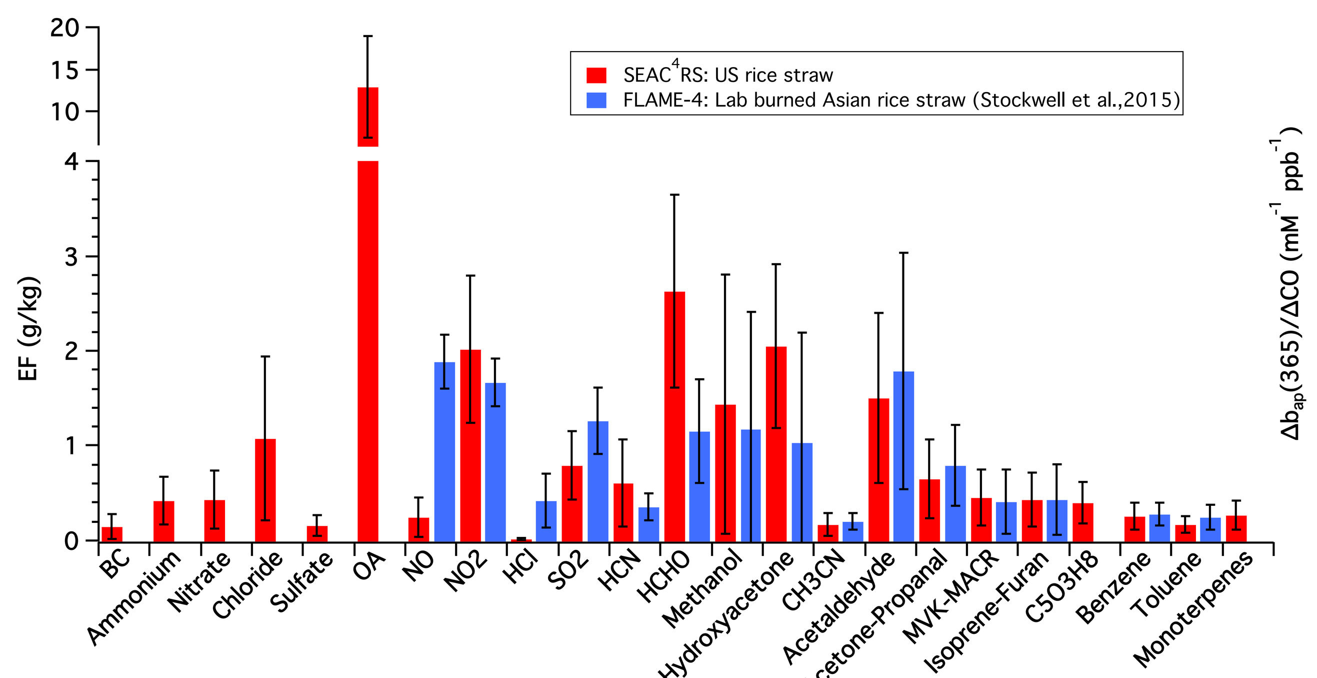


Figure 2. Emission factors (g kg^{-1}) measured during SEAC[4]RS and FLAME-4.

- In general, the average EFs of SEAC[4]RS and those predicted from the FLAME-4 EF vs. MCE plot for the overlapping species are shown to agree well

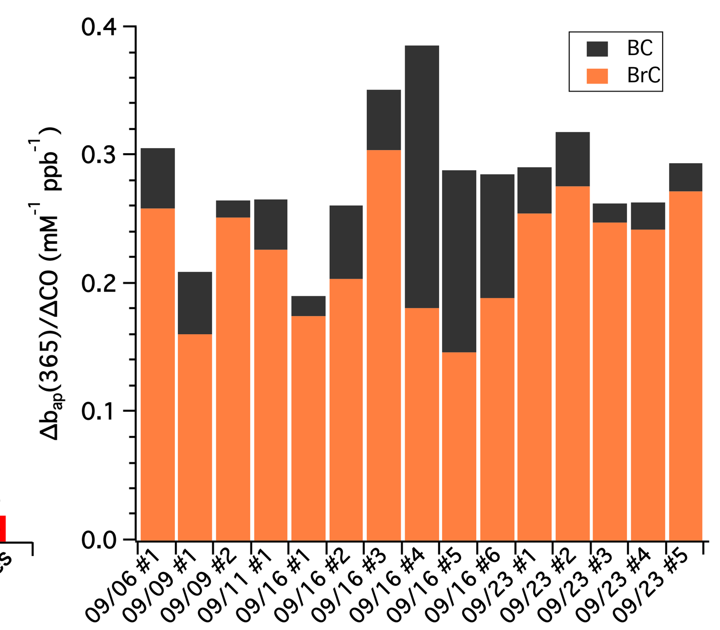
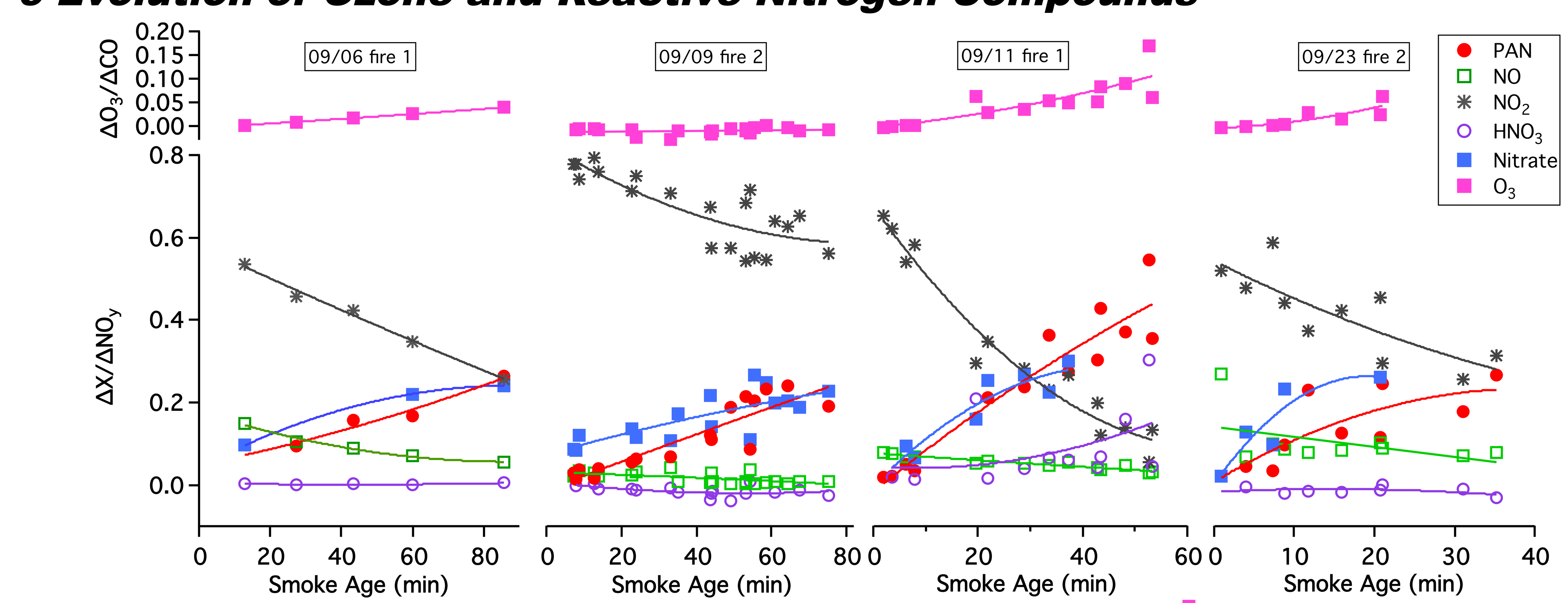
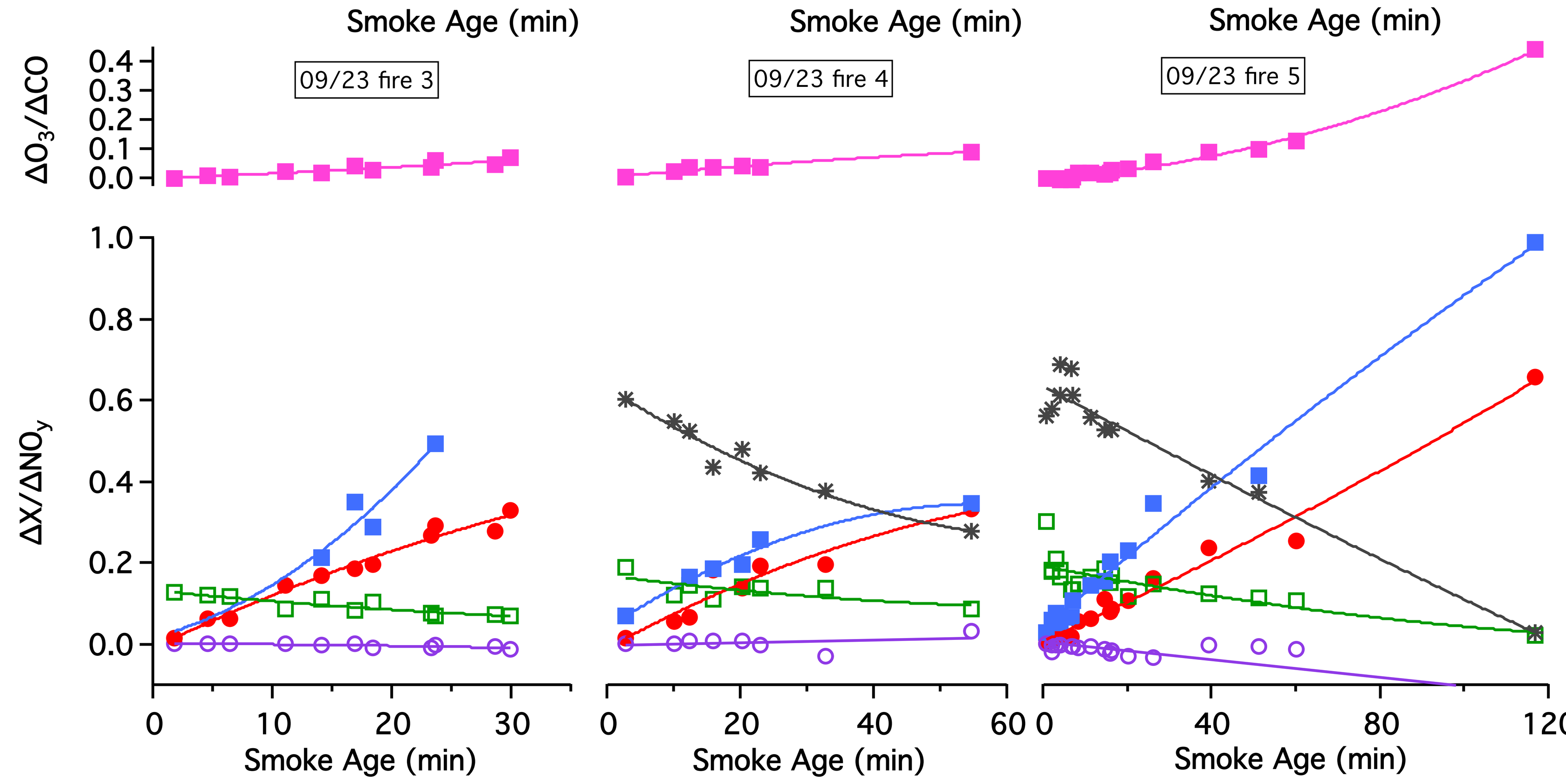


Figure 3. Total light absorption coefficient at 365 nm with BrC and BC contributions.

- Absorption Ångström Exponent (AAE) inferred from PSAP absorption at 470 and 532 nm
- BrC absorption at 365 nm derived from aerosol AAE and a BC AAE of 1
- PSAP data indicate that BrC is ubiquitous in agricultural fire plumes

6 Evolution of Ozone and Reactive Nitrogen Compounds





- Rapid increase in $\Delta O_3/\Delta CO$ observed in aging plumes, up to ~10% in 1 h.
- NO_x was rapidly converted to PAN and particulate nitrate
- HNO_3 showed no enhancement, likely converted to nitrate due to NH_3 in plumes
- Variability in initial emissions and photochemical processing affect smoke chemistry

Figure 4. Evolution of NO_y species and O_3 in 7 aging plumes .

7 Evolution of OA and BrC

- No significant difference downwind in the first hour of aging

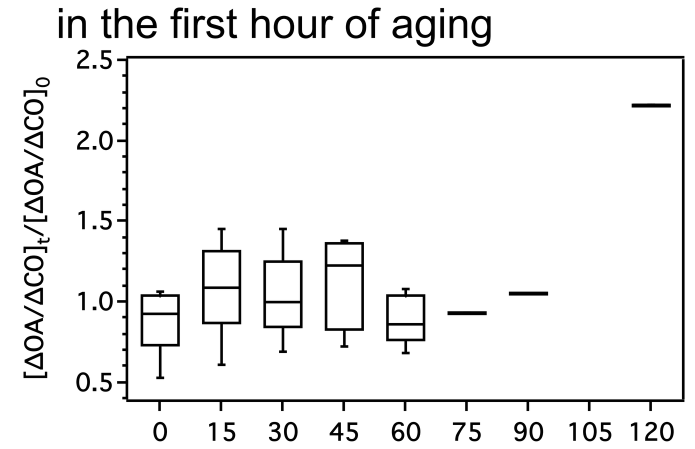


Figure 5. Whisker plot of OA evolution.

- Increase in O:C and decrease in H:C observed in some fires

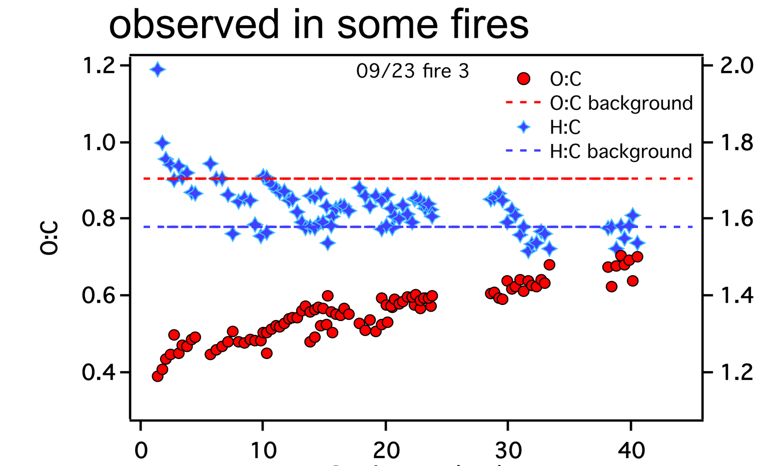


Figure 6. Change of O:C and H:C ratios.

- Very slight increase in BrC absorption

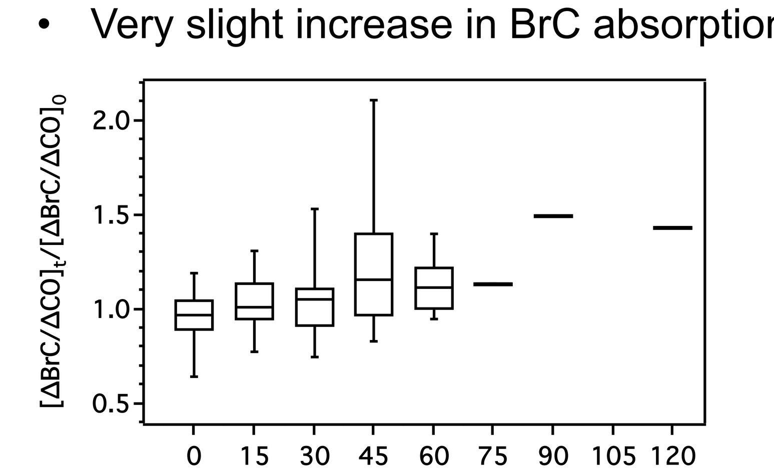
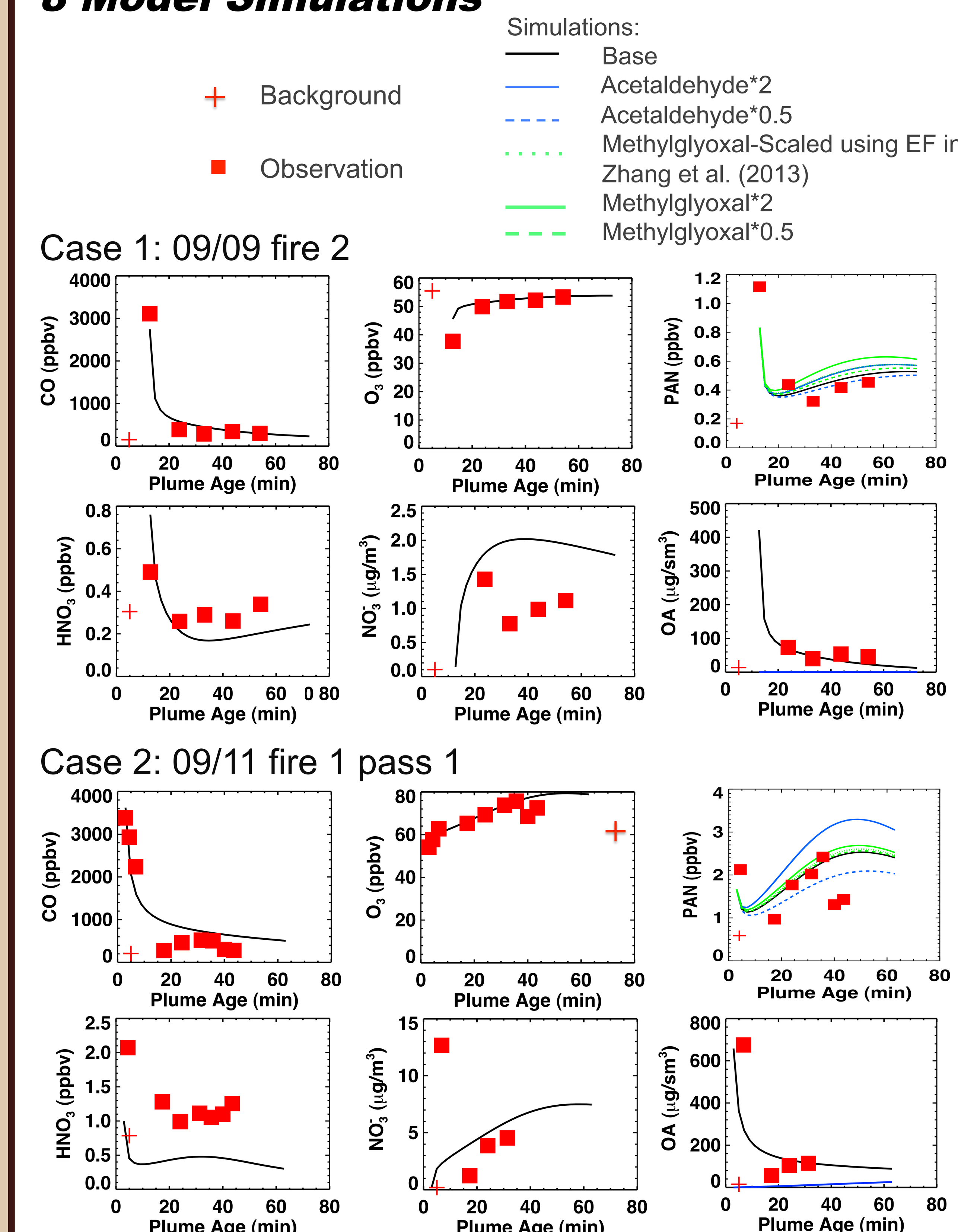


Figure 7. Whisker plot of BrC abs evolution.

8 Model Simulations



- Modeled O_3 and PAN agree with the observations within ~30%.
- PAN is sensitive to OVOCs, e.g., acetaldehyde and methylglyoxal.
- The HNO_3-NO_3^- system is more complicated than the model can represent. For example, while the model assumes no initial emission of nitrate, AMS data show the opposite for Case 2.
- Modeled SOA production is small compared to the OA variability.

9 Conclusions

- Emission factors for a set of gaseous and particulate species were measured.
- BrC was produced in agricultural fires.
- As the plumes aged, fast enhancement of O_3 and conversion of NO_x to PAN and nitrate were observed.
- The ratio of OA to CO and that of $b_{ap,\,BrC}$ to CO were not significantly different downwind during the first hour of aging. However, the increase of O:C ratio indicates ongoing photo-oxidation.
- The model reasonably simulates dilution and chemical processes in aged fire plumes, especially for O_3 and PAN chemistry.

Acknowledgement

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