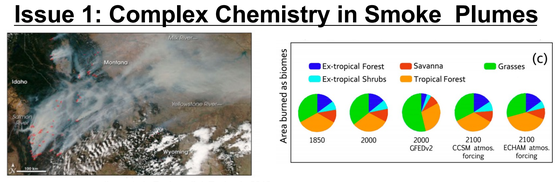
Adding On-Line Photolysis Calculations to a 2D Lagrangian Model of Smoke Plume Chemistry to Investigate In-Plume Gradients

Matthew J. Alvarado (malvarad@aer.com), Marikate Mountain, Archana Dayalu

Motivation

Issue 1: Complex Chemistry in Smoke Plumes

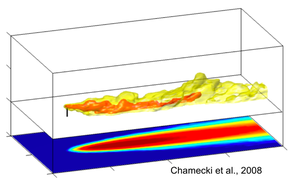


Jiang et al., Environ. Sci. Technol., 2012

Emissions are highly variable between fires and fire types. Rapid near-source chemistry creates O_3, PAN, SOA, etc.

Understanding this chemistry is critical to assessing air quality, health and climate impacts from biomass burning.

Issue 2: Plume Chemistry is at Sub-grid Scales



Chamecki et al., 2008

Global and regional CTMs can unphysically "mix" emissions within the large-scale grid boxes.

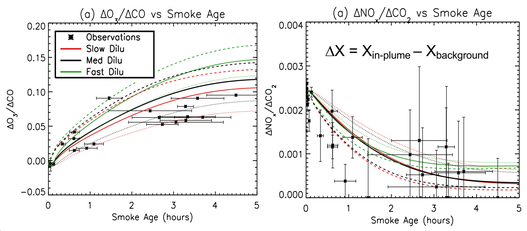
This can lead incorrect estimates of biomass burning impacts.

Plume-scale process models allow us to:

- Examine the chemical transformations within the smoke plumes
- Develop parameterizations of aging process for coarser grid-scale CTMs

Models

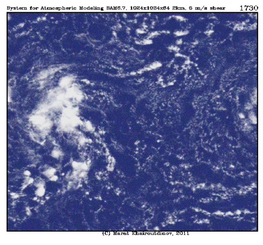
Aerosol Simulation Program (ASP v2.1)



- Gas-phase chemistry
  - ≤C_4 gases follow Leeds Master Chemical Mechanism v3.2
  - Other organic gases follow RACM2 (Goliff et al., 2013)
- OA thermodynamics using the Volatility Basis Set (VBS) (Robinson et al., 2007)
- **Adding reasonable SVOC chemistry can simulate OA, O_3, OH, and NO_x observations (Alvarado et al., ACP, 2015).**

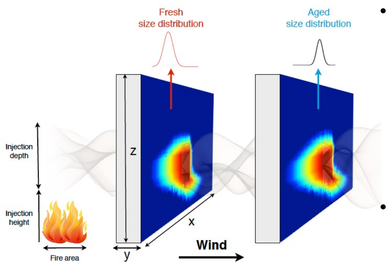
System for Atmospheric Modeling (SAM)

- Non-hydrostatic Large Eddy Simulation (LES) model
- Several microphysics packages
- 1.5-order sub-grid scale closure (prognostic SGS TKE) or Smagorinsky-type closure
- Radiation from AER's RRTM
- Available at http://rossby.msrc.sunysb.edu/%7Emarat/SAM.html



Khairoutdinov and Randall, 2003

Coupling SAM to ASP: SAM-ASP v1.0

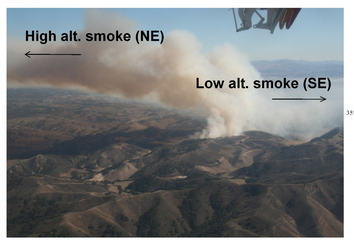


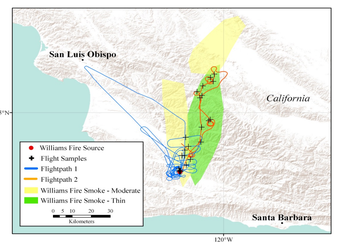
Approach based on SAM-TOMAS (Sakamoto et al., ACP 2015, 2016)

- **SAM**
  - Calculates transport of all species as 2D Lagrangian Wall
  - Calculates the temperature, pressure, air density, and solar zenith angle
- **ASP**
  - Calculates the gas and aerosol processes for each grid box
  - Photolysis rates are calculated using ***off-line*** look-up tables

SAM-ASP v1.0 Evaluation (Lonsdale et al., GMD, 2020)

SAM-ASP Test Case: Williams Fire (Akagi et al., 2012)

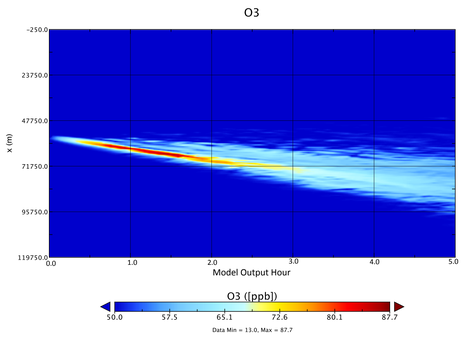




- **The Williams Fire** (burning scrublands) was sampled from 10:50-15:20 LT on Nov. 17, 2009. Skies were clear with low RH and variable winds (2-5 m/s).
- Measurements included **U. Montana airborne FTIR** (CO, O_3, NO_x, PAN, etc.), **compact ToF-AMS** (OA), **SP2** (BC), **nephelometer**, and meteorological data.
- Significant chemical formation of O_3 and PAN, but slight *loss* of OA downwind!

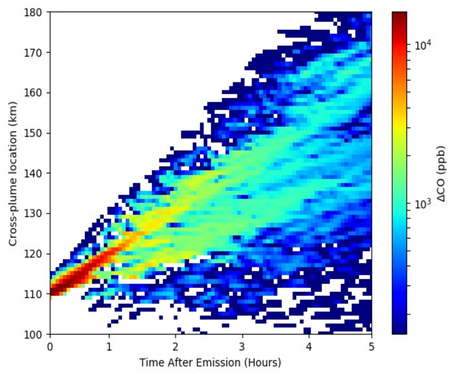
SAM-ASP Model Setup

- Domain (x,z) = 120 km x 3km
- Gridsize (x,z) = 500 m x 40 m
- Meteorology is driven by nudging and boundary conditions from assimilated meteorology from the National Center for Environmental Prediction (NCEP) North American Regional Reanalysis (NARR) data (Mesinger et al., 2006).
- Emissions of CO scaled to match observed initial CO concentration
- Emissions of all other species determined from measured or literature estimates of emissions ratios to CO



Plume Dilution and Transport

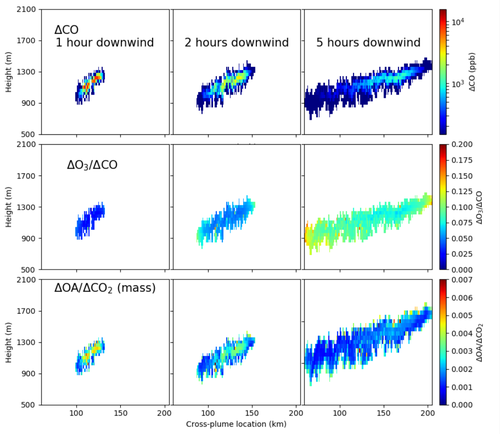
- Significant dilution of CO in plume, as expected.
- Dilution in first hour is slower than in Alvarado et al. (2015), which used a parameterized fit to dilution of a Lagrangian box
- But concentrations at > 1 hour downwind are consistent with Lagrangian box approach
- Plume approximately 80 km wide 5 hours downwind, average ΔCO of ~1000 ppbv.



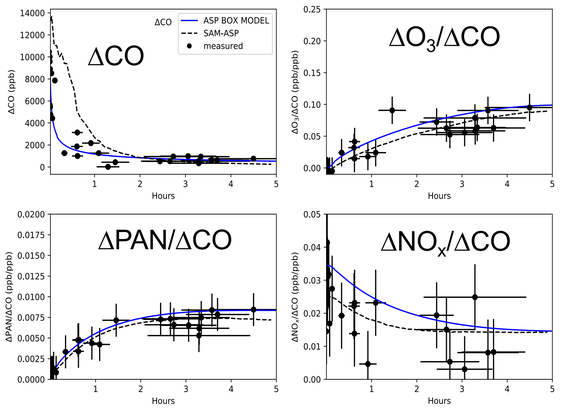
Top-down view at injection height (1.2 km agl)

Gradients within the Smoke Plume

- Clear horizontal and vertical gradients in ΔCO (top row)
- $\Delta O_3/\Delta CO$ (middle row) slightly lower in core of plume, likely due to NO titration of O_3
- $\Delta OA/\Delta CO_2$ (bottom row) higher in core of plume.



Comparison to ASP Box Model Results: Gases

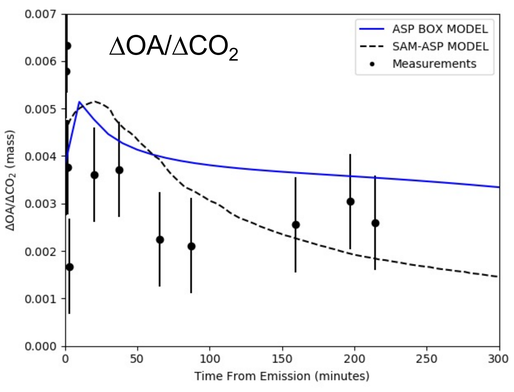


Average all SAM-ASP boxes with CO > 150 ppb for comparison.

As already noted, dilution in first hour is slower than in box study

However, formation of O_3 and PAN, as well as loss of NO_x, is consistent with observation and box model results.

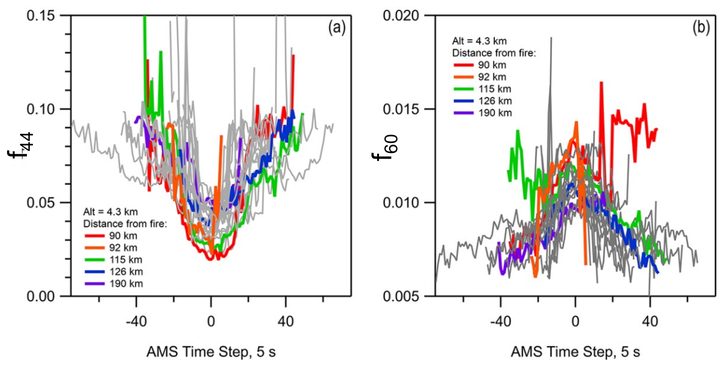
Comparison to ASP Box Model Results: OA



- Within the first hour after emission, SAM-ASP has less dilution and evaporation than the box model.
- However, SAM-ASP has greater dilution than the box model after 2 hours, which lead to more OA evaporation in SAM-ASP than in the box model, leading to a lower OA NEMR after 2 hours.

Current Work: Online Photolysis Rate Calculations with TUV

Issue 3: Simulating Horizontal and Vertical Gradients in Photolysis and Chemistry

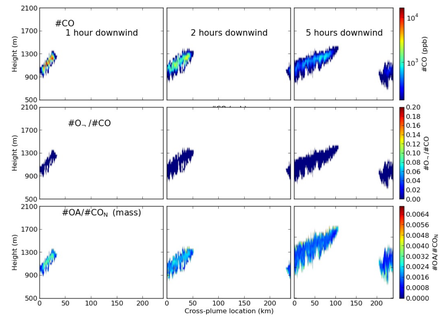


AMS f44 and f60 OA fragments for cross-plume transects from the South Sugarloaf fire during WE-CAN. From Garofalo et al. (2019).

- f_{60}, a marker for fresh BB OA, is depleted at the edges of the plume relative to the core.
- f44, a marker for oxidized OA, is enhanced at the plume edges relative to the core.
- $\Delta OA/\Delta CO_2$ somewhat more enhanced at the plume core than the edges (not shown).

Calculating Photolysis Rates On-line using TUV

- We have added TUV as an on-line subroutine to SAM-ASP
- TUV uses ASP-calculated aerosol optical properties (Alvarado et al., ACP, 2016) to calculate photolysis rates
- Current version gives too dark a plume (little O_3 formation, see below), and we are investigating if the problem is on the smoke optical properties or on the model coupling.



Acknowledgements and funding agencies:

NSF Atmospheric Chemistry Grant #1558966

NOAA CPO Atmospheric Chemistry Carbon Cycle and Climate (AC4) Grant #NA17OAR4310002