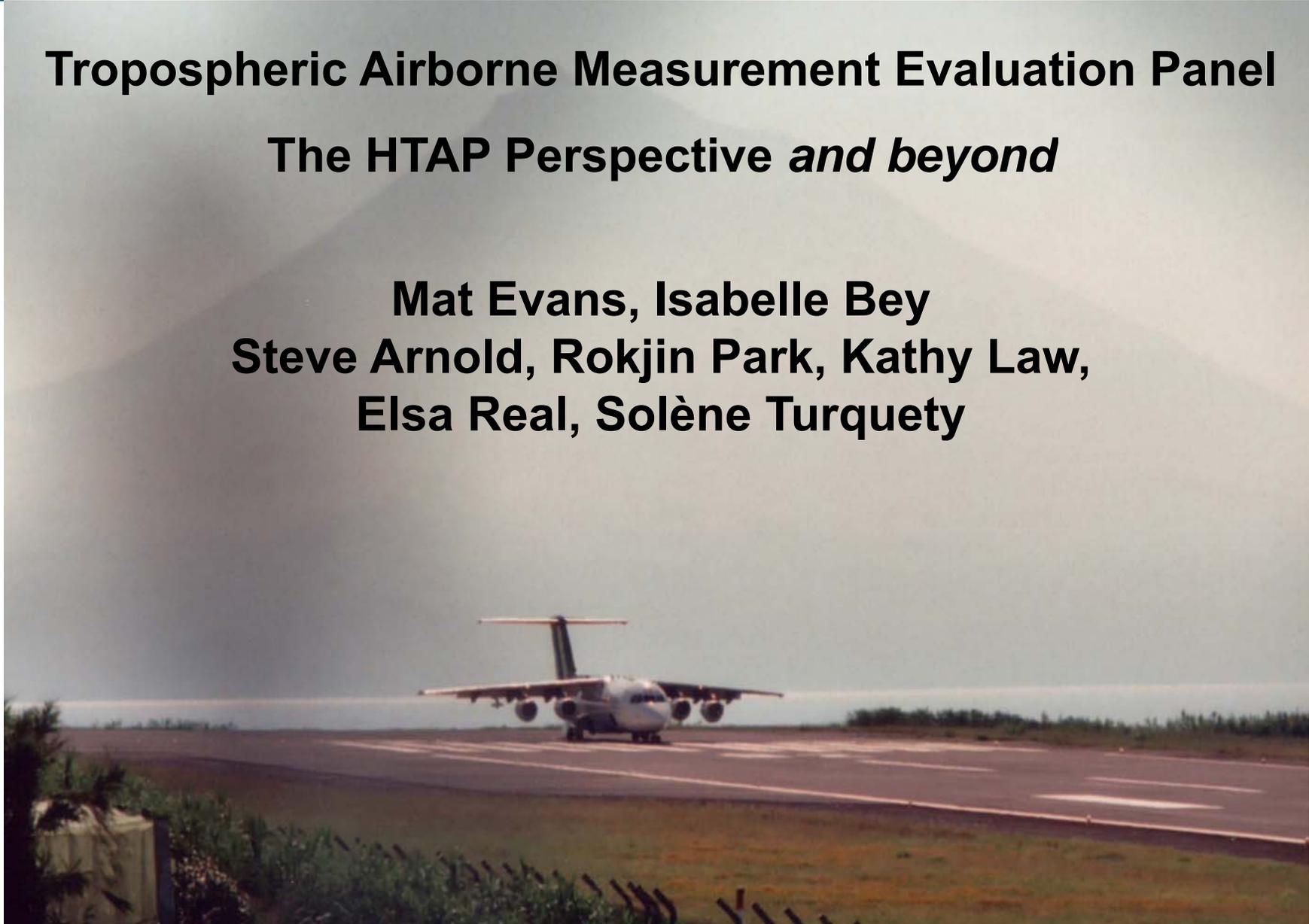


Tropospheric Airborne Measurement Evaluation Panel

The HTAP Perspective *and beyond*

**Mat Evans, Isabelle Bey
Steve Arnold, Rokjin Park, Kathy Law,
Elsa Real, Solène Turquety**



What is HTAP?

To develop a fuller understanding of intercontinental transport of air pollution in the Northern Hemisphere, the Executive Body of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention) established the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) to:

- (a) Plan and conduct the technical work necessary to develop a fuller understanding of the hemispheric transport of air pollution for consideration in the reviews of protocols to the Convention;
- (b) Plan and conduct the technical work necessary to estimate the hemispheric transport of specific air pollutants for the use in reviews of protocols to the Convention and prepare technical reviews thereon for submission to the Steering Body of EMEP;
- (c) Carry out such other tasks related to the above work as the Executive Body may assign to it in the annual work-plan. [See Annex IV of ECE/EB.AIR/83/Add.1]

HTAP Experiment Set 3 Event Simulation

➤ Objectives

Evaluate and intercompare (some of) the models contributing to HTAP with respects to their capabilities to reproduce the long-range transport of pollution using the ICARTT data set

➤ Activities proposed by:

M. Evans, R. Park, I. Bey, S. Turquety, K. Law, E. Real, S Arnold,

A rather Harvard Mafia! ☺

HTAP Experiment Set 3 – time line, 1

Requested simulations

- **ES1.** A standard simulation for 2004 with specified biomass burning inventory (taken from Turquety et al., [2007]) and injection height. Model outputs requested over the period from June to September 2004.
- **ES2.** A sensitivity simulation with North American anthropogenic emissions reduced by 20% from March 1st to September 30th 2004.
- **ES3.** A sensitivity simulation with North American biomass burning emissions reduced by 20% from March 1st to September 30th 2004 over the region defined in the Turquety et al., files.
- **ES4.** A sensitivity simulation similar to ES1 with biomass burning emissions restricted to the boundary layer from May 1st 2004 onward.

Requested diagnostics (monthly mean + 3-hour timeseries)

- Trace gas concentrations
- Aerosol concentrations
- Aerosol optical depths
- Deposition rates
- Chemical tendencies (ozone and CO)
- Emissions
- Meteorological data (pres, temp, convective mass fluxes)
- Photolysis rates

- **General characterisation of chemical signatures of different air masses over the North Atlantic area**
- **Comparison model outflow characteristics**
- **Aerosol export (export efficiency of black carbon aerosols)**
- **Impact of injection height on long range transport of biomass burning emissions**
- **On-route processing of plumes of biomass burning and anthropogenic origins**

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ICARTT

We collected a lot of observations during a field campaign.

They are usually get 'sliced and diced' by 'experience'

But can we classify them systematically minimizing the prior assumption?

Can we then use this classification to assess our understanding of the processes occurring?

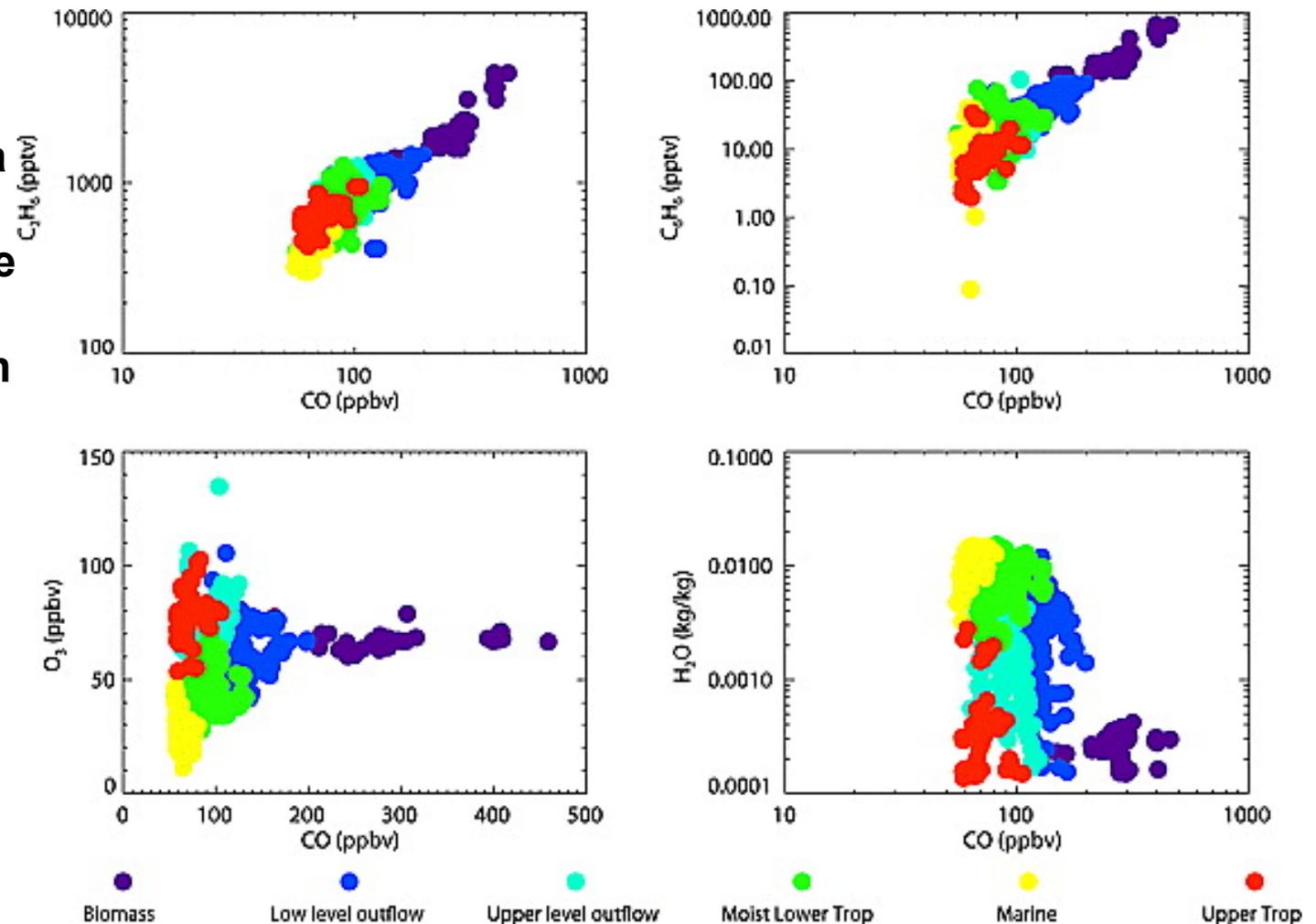
Cluster analysis

allows the partitioning of a data set into subsets (clusters), so that the data in each subset share some common trait.

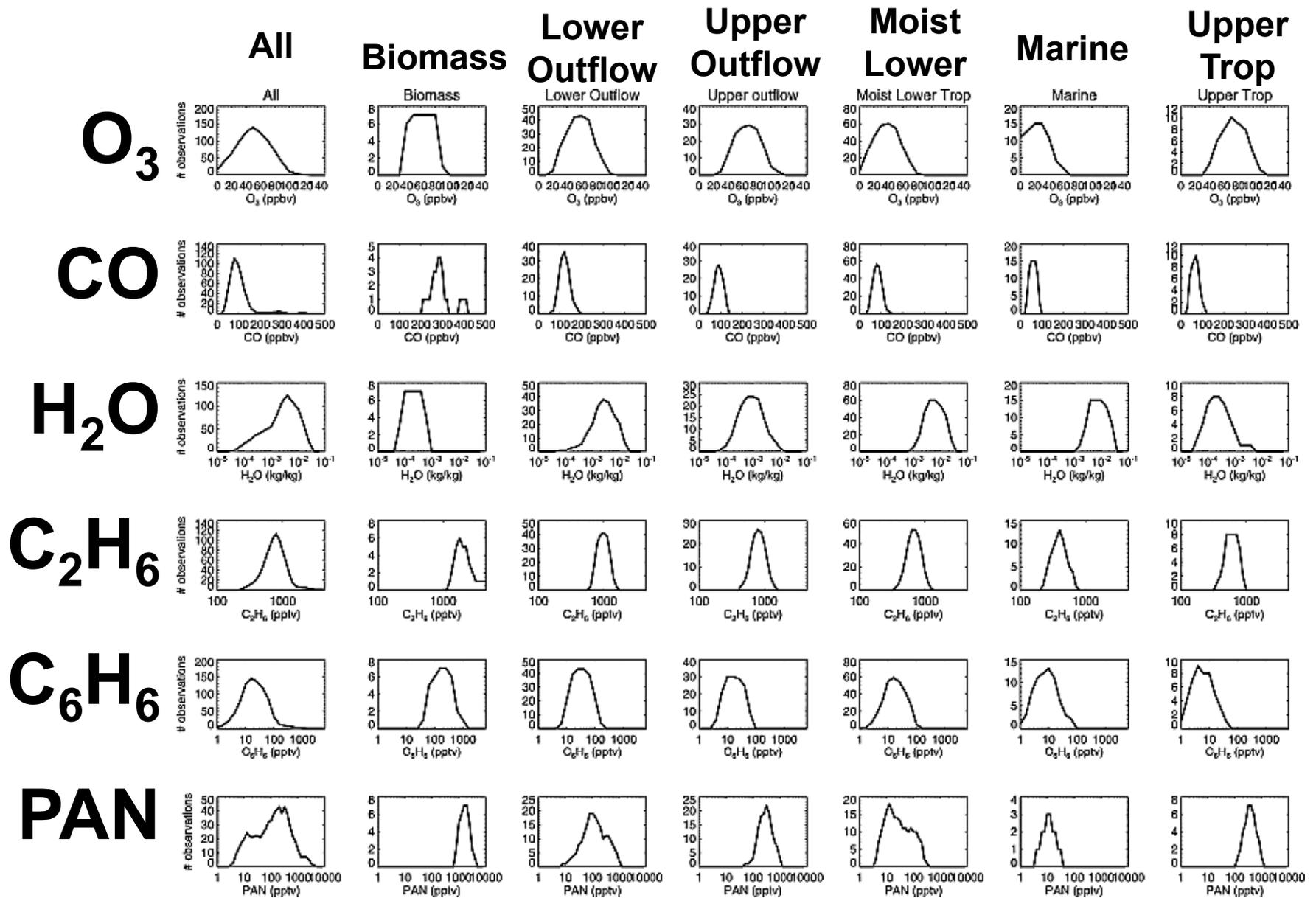
In this case we used

- $[O_3]$
- $\log(q)$
- $[C_6H_6]$
- $[C_2H_6]$

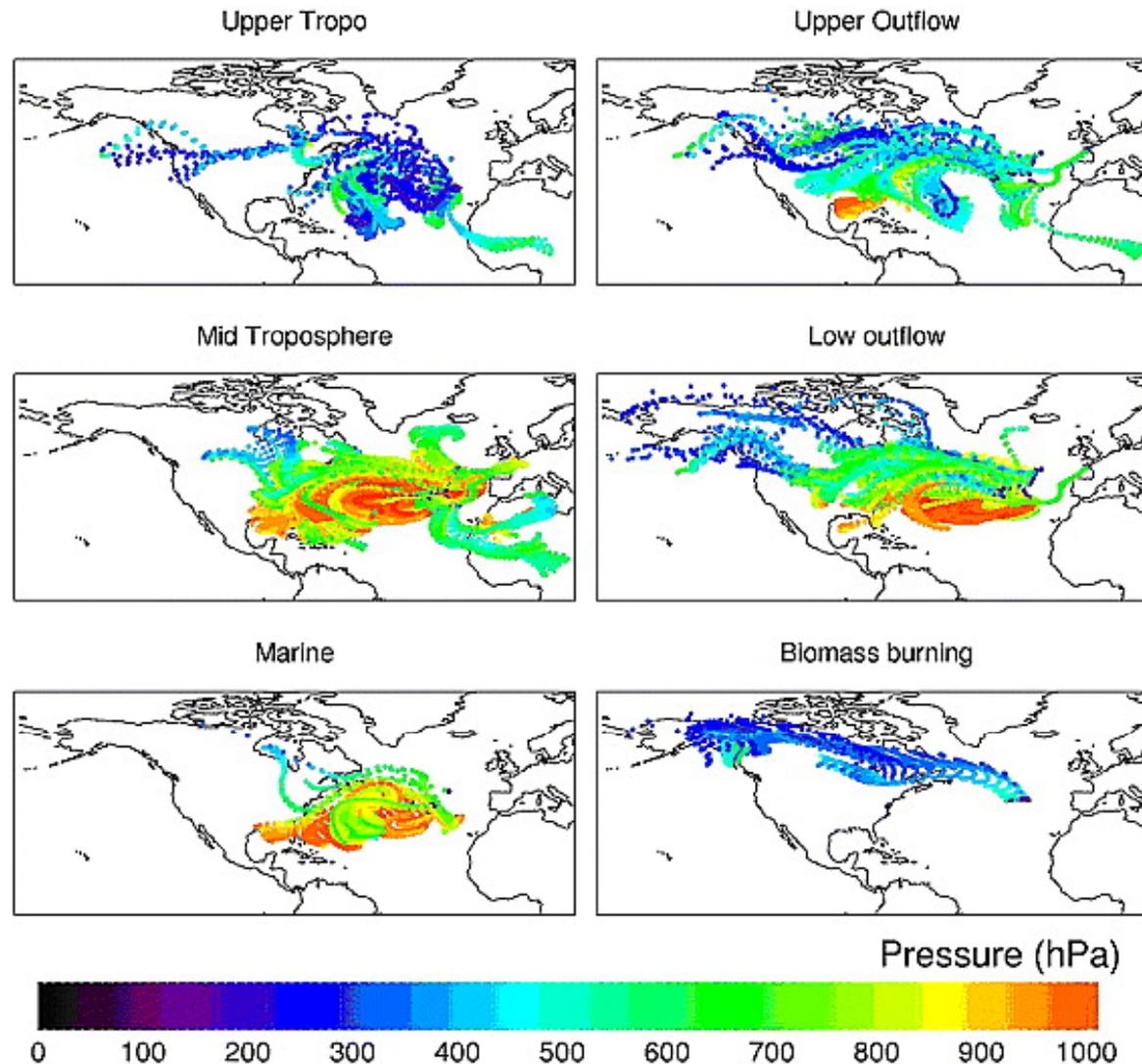
from the BAe146 during ICARTT



A.C. Lewis, M.J. Evans, et al., Chemical composition observed over the mid-Atlantic and the detection of pollution signatures far from source regions, *J. Geophys. Res.*, 2007.

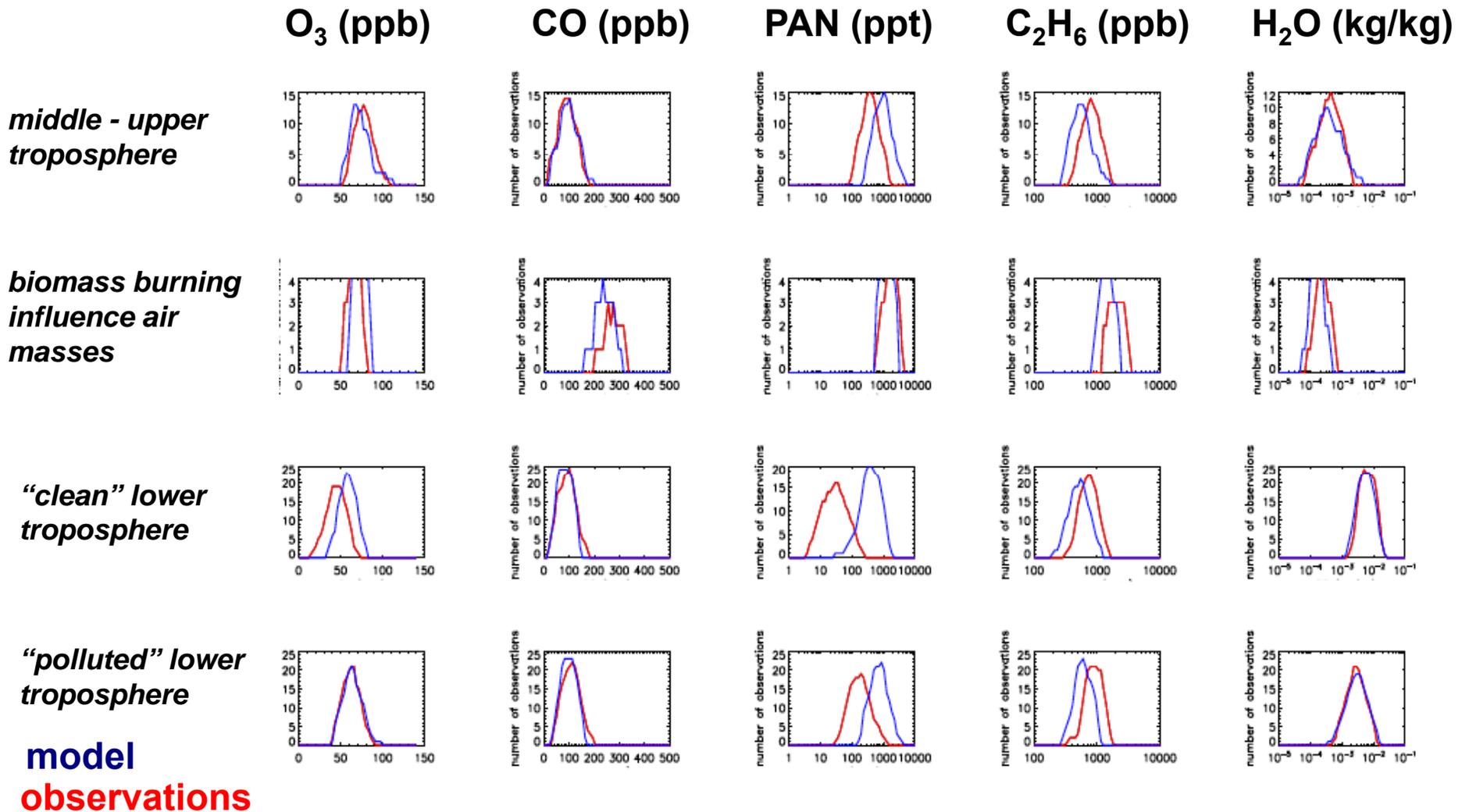


Do the clusters tie up with the meteorology (trajectories)?



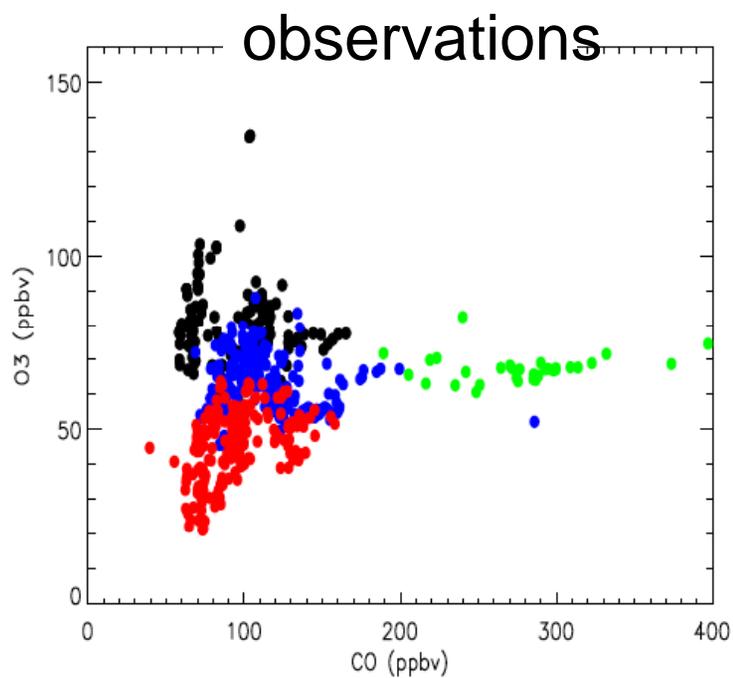
Can we use these approaches to test models?

- Are the characteristic air masses (as manifested by the clusters) in the models the same as those observed?
- Is the composition of the clusters comparable between the models and between models and observations?
- In which clusters is the model failure most significant? Can we attribute this failure to a particular model process?



Preliminary results from the French model MOCAGE,
courtesy of N. Boussez and J.-L- Attié, Laboratoire d'aérodologie, Toulouse, France

Within each cluster are the relationships species the same?
Principal components analysis will allow us to investigate this

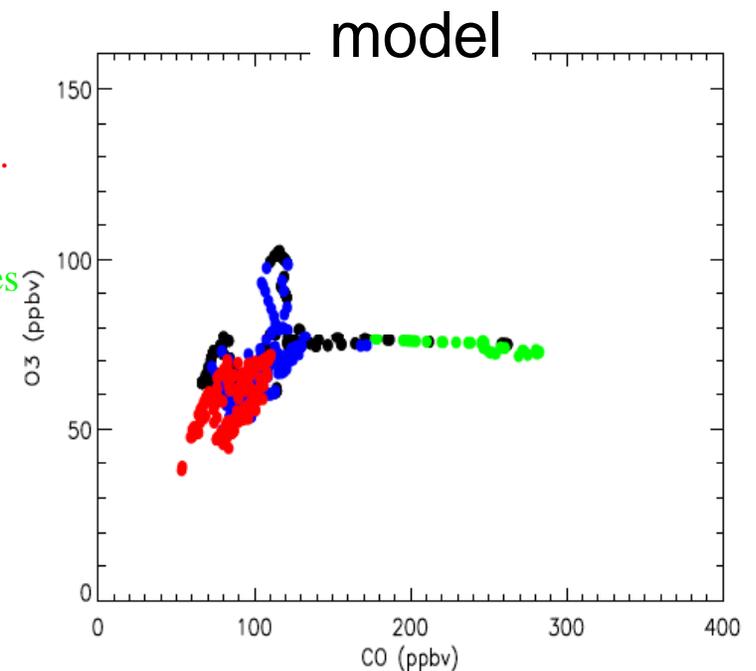


“clean” lower trop.

biomass burning
influenced air masses

middle-upper
troposphere

“polluted” lower
trop.



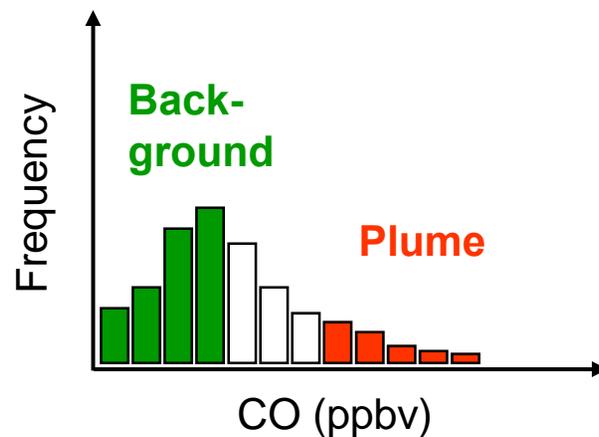
- **General characterisation of chemical signatures of different air masses over the North Atlantic area**
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- **On-route processing of plumes of biomass burning and anthropogenic origins**

Objective:

Examine in a quantitative manner the overall impact of plumes on the O_3 production on specific regions such as e.g. North Atlantic and intercompare different models

Methodology:

- Differentiate the “polluted” and “background” environments (e.g. identify the ensemble of plumes in the 3D fields) using various criteria (e.g. ΔCO , ΔNO_x)
- Examine the characteristics (O_3 tendencies, water vapor, etc.) of the ensemble of plumes



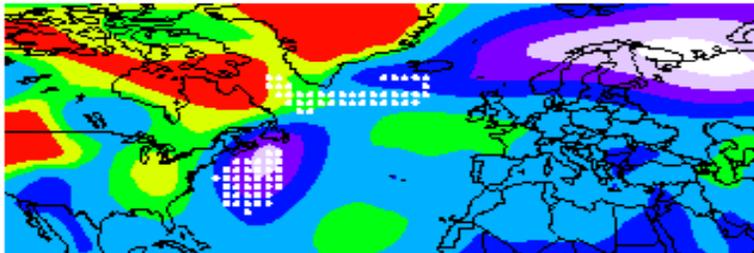
Plume: $CO_{daily} > (\text{median} + \text{standard deviation}) CO_{monthly}$

Background: $CO_{daily} < (\text{median}) CO_{monthly}$

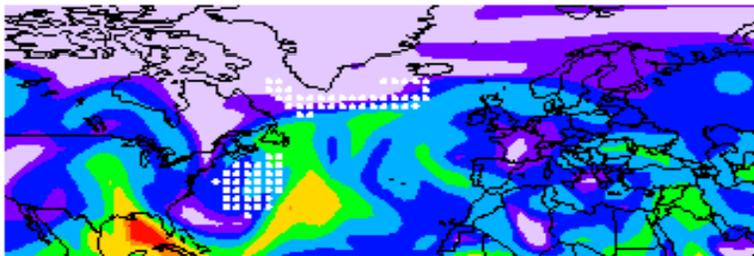
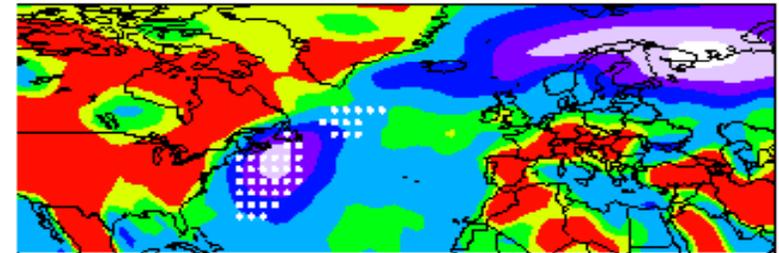
GEOS-Chem

02/04/1997

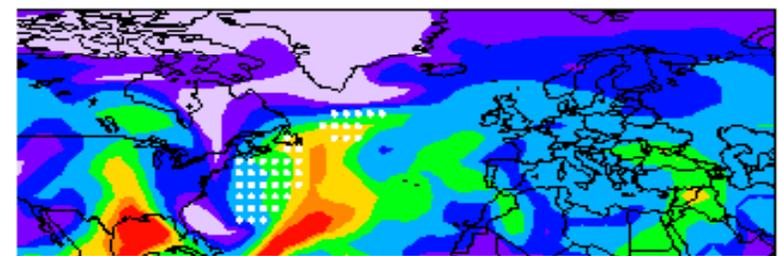
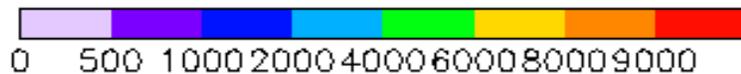
ECHAM5-MOZ



Mean sea level
pressure (hPa)

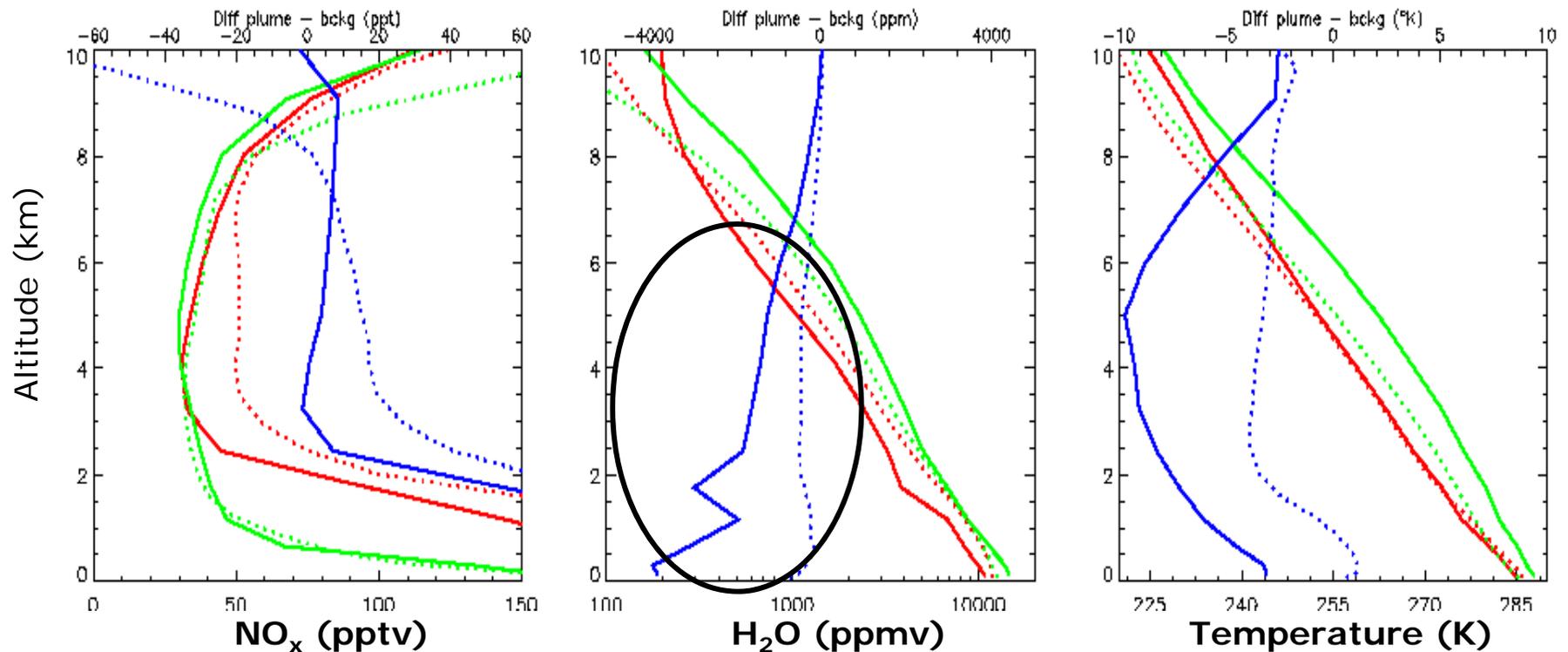


H₂O (ppmv)
at 700 hPa



Impact of North American
outflow over the North
Atlantic – April 1997

- GEOS-Chem Background
- ⋯ MOZECH Background
- GEOS-Chem Polluted
- ⋯ MOZECH Polluted
- GEOS-Chem polluted-background
- ⋯ MOZECH polluted-background



- **General characterisation of chemical signatures of different air masses over the North Atlantic area**
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$$f_X(z) = \frac{1}{R_X} \left(\frac{\Delta[X]}{\Delta[CO]} \right) (z)$$

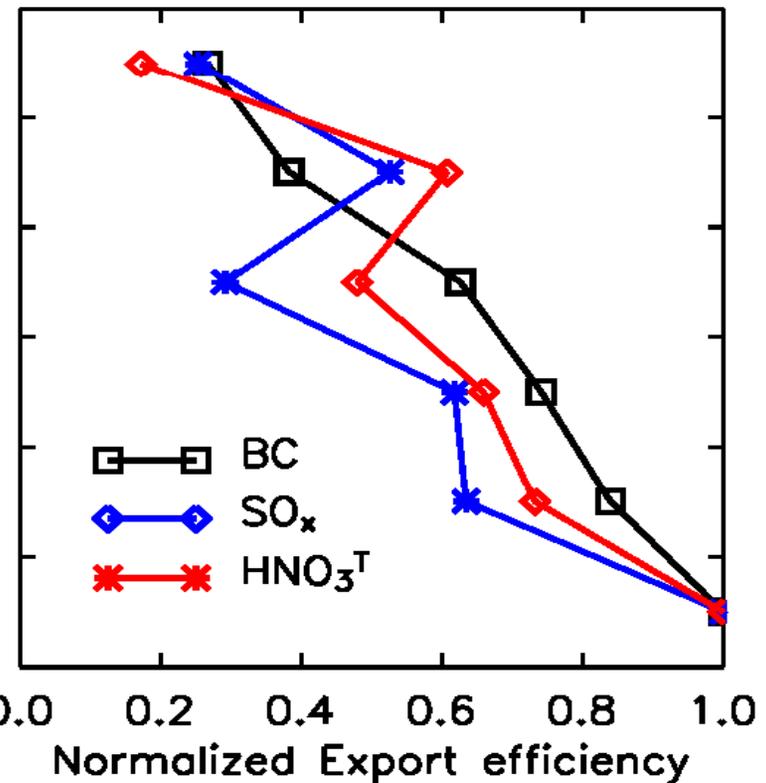
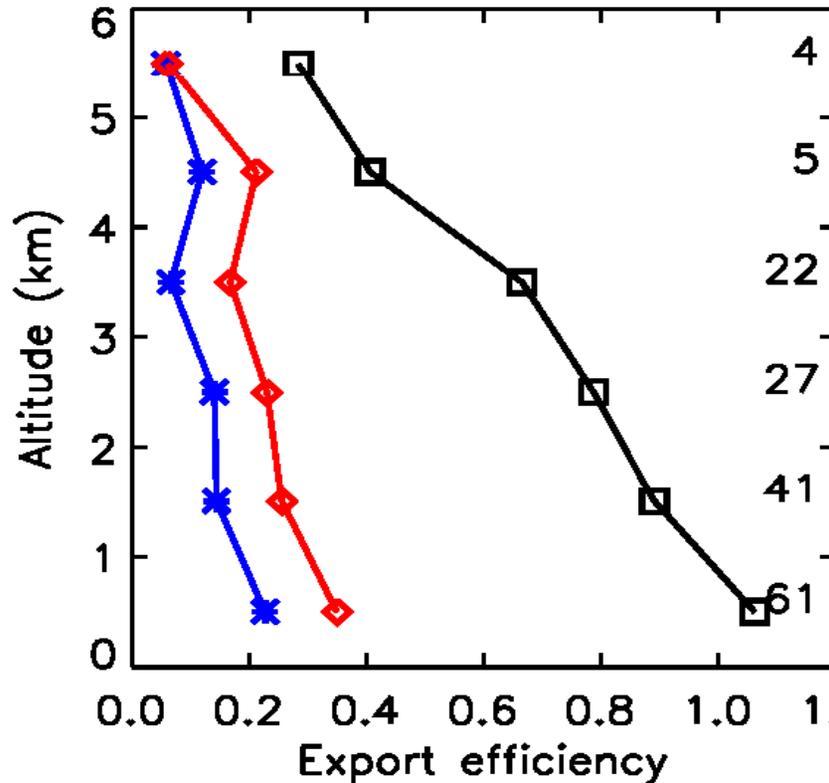
$$f_{X \text{ norm}}(z) = \frac{f_X(z)}{f_X(0)}$$

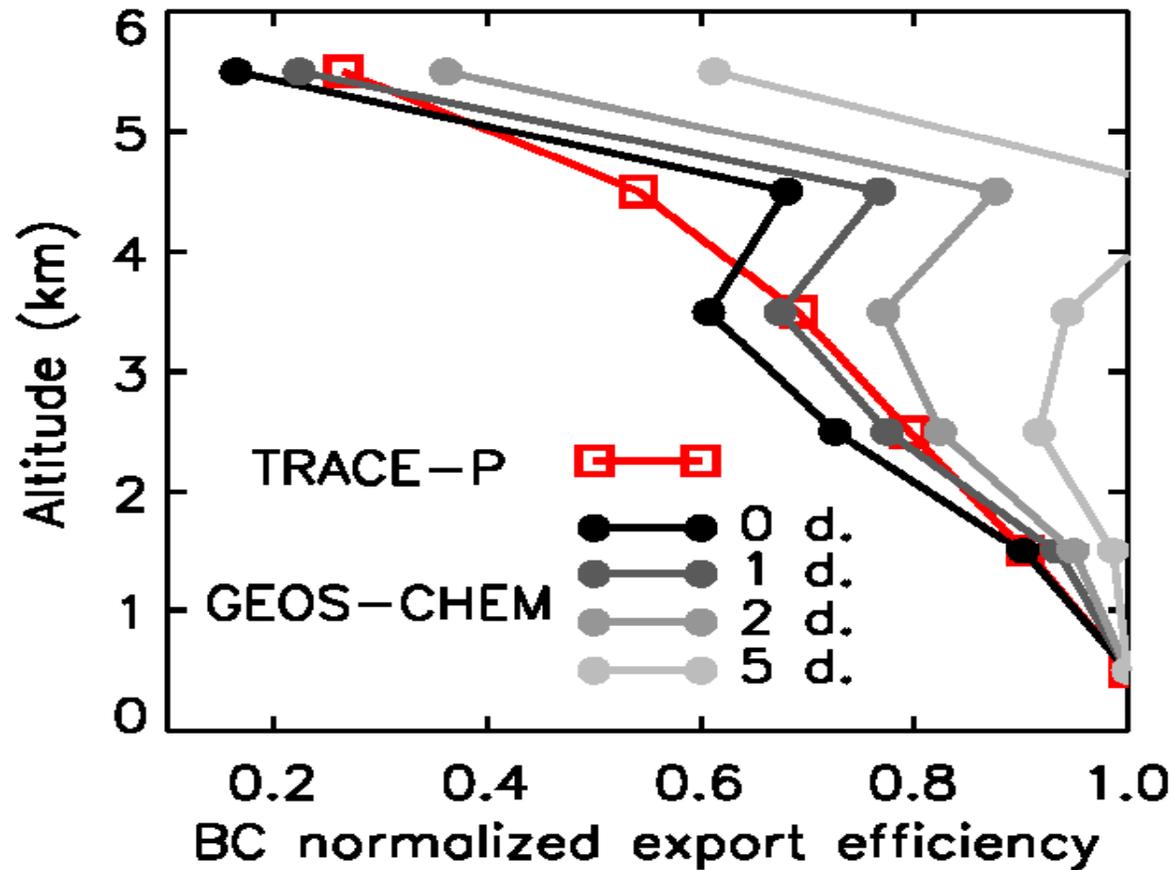
[Koike et al., 2003;
Parrish et al., 2004]

X = combustion-derived species

R_X = emission ratio (X/CO)

Δ = enhancements relative to background



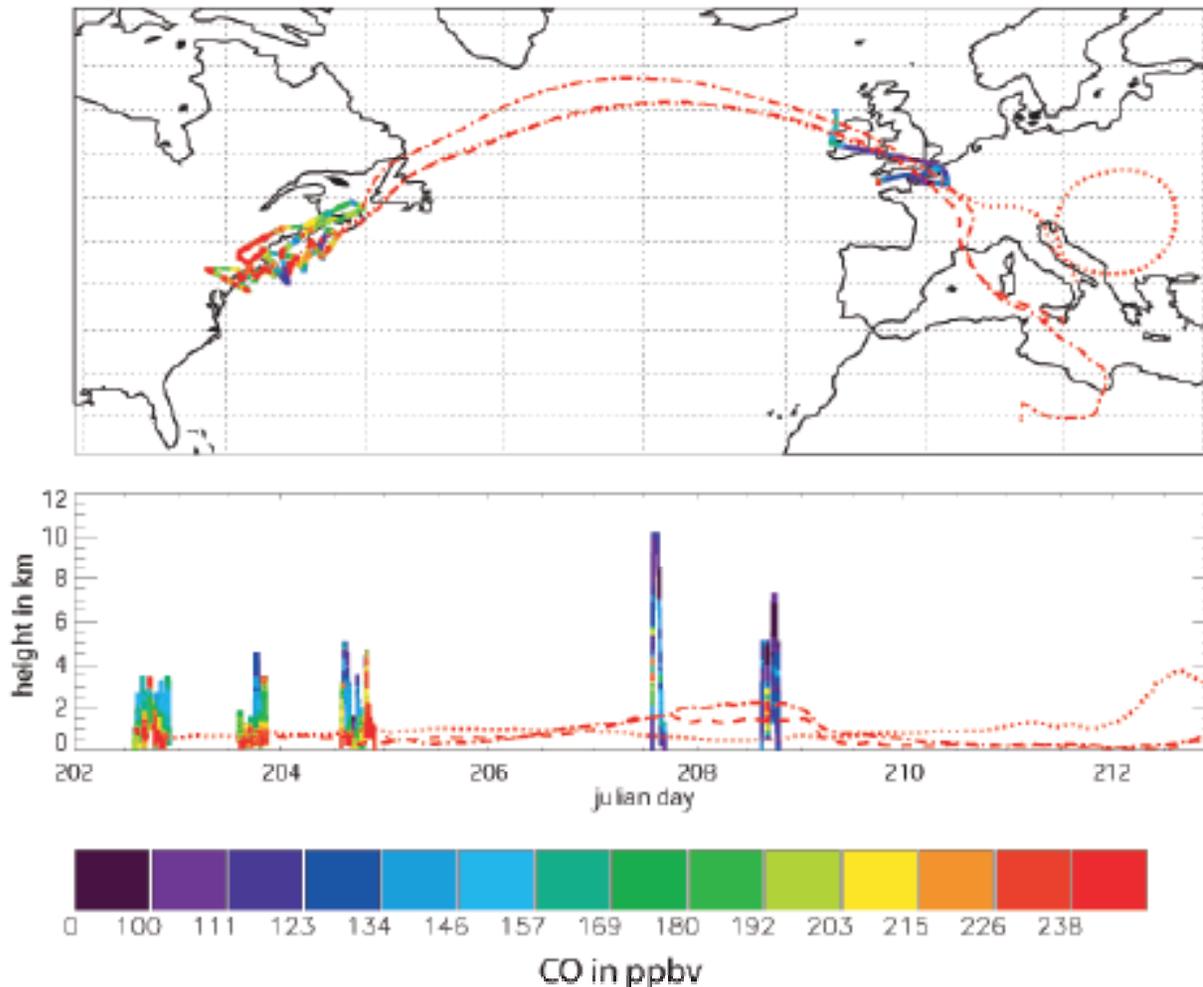


timescale τ for conversion of hydrophobic to hydrophilic BC in global models

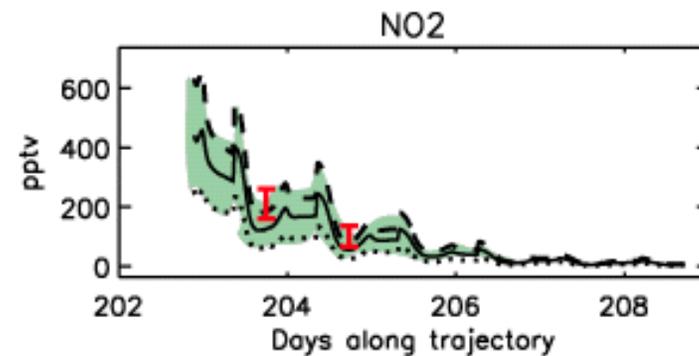
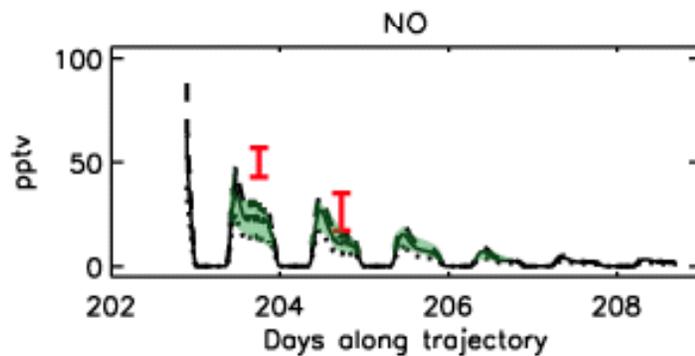
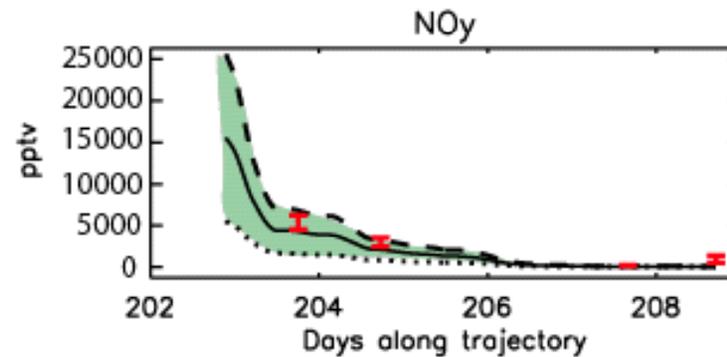
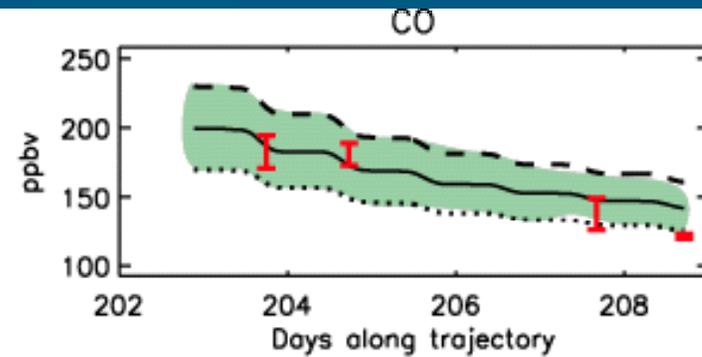
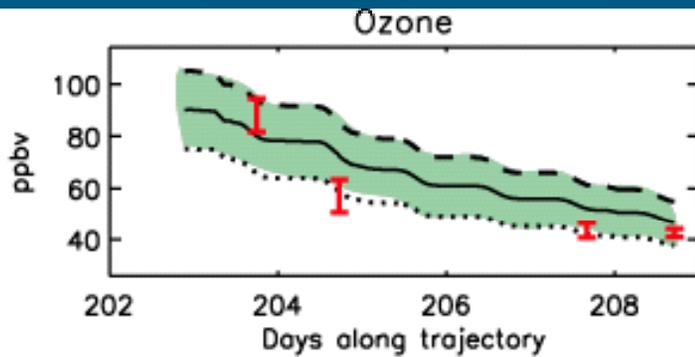
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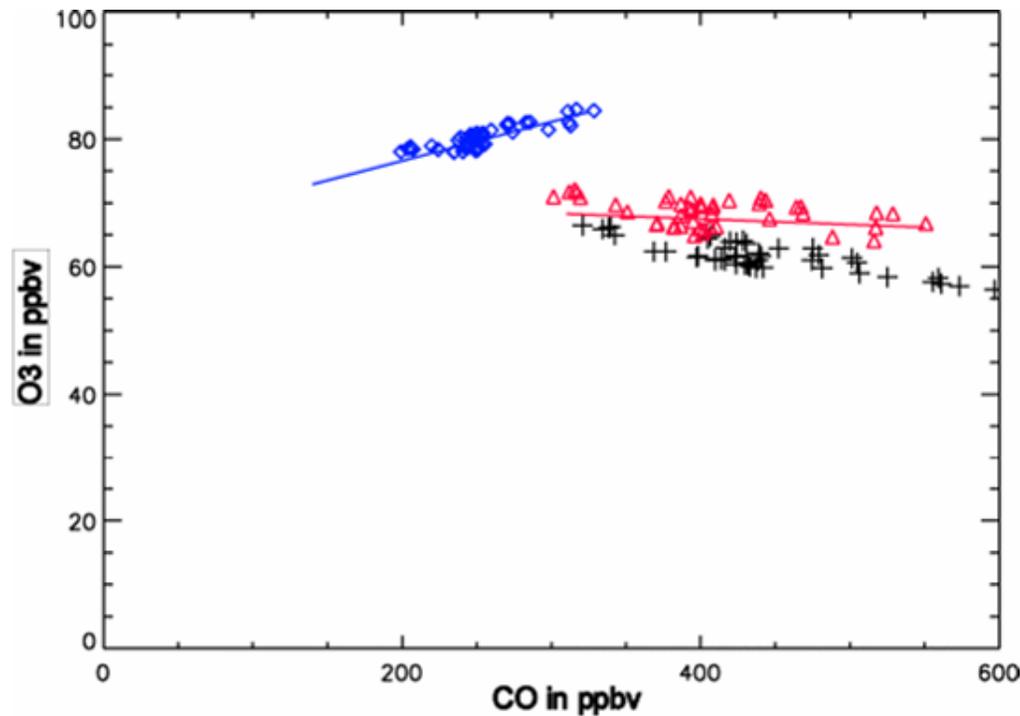
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Kathy Law + Steve Arnold



Real et al., 2008



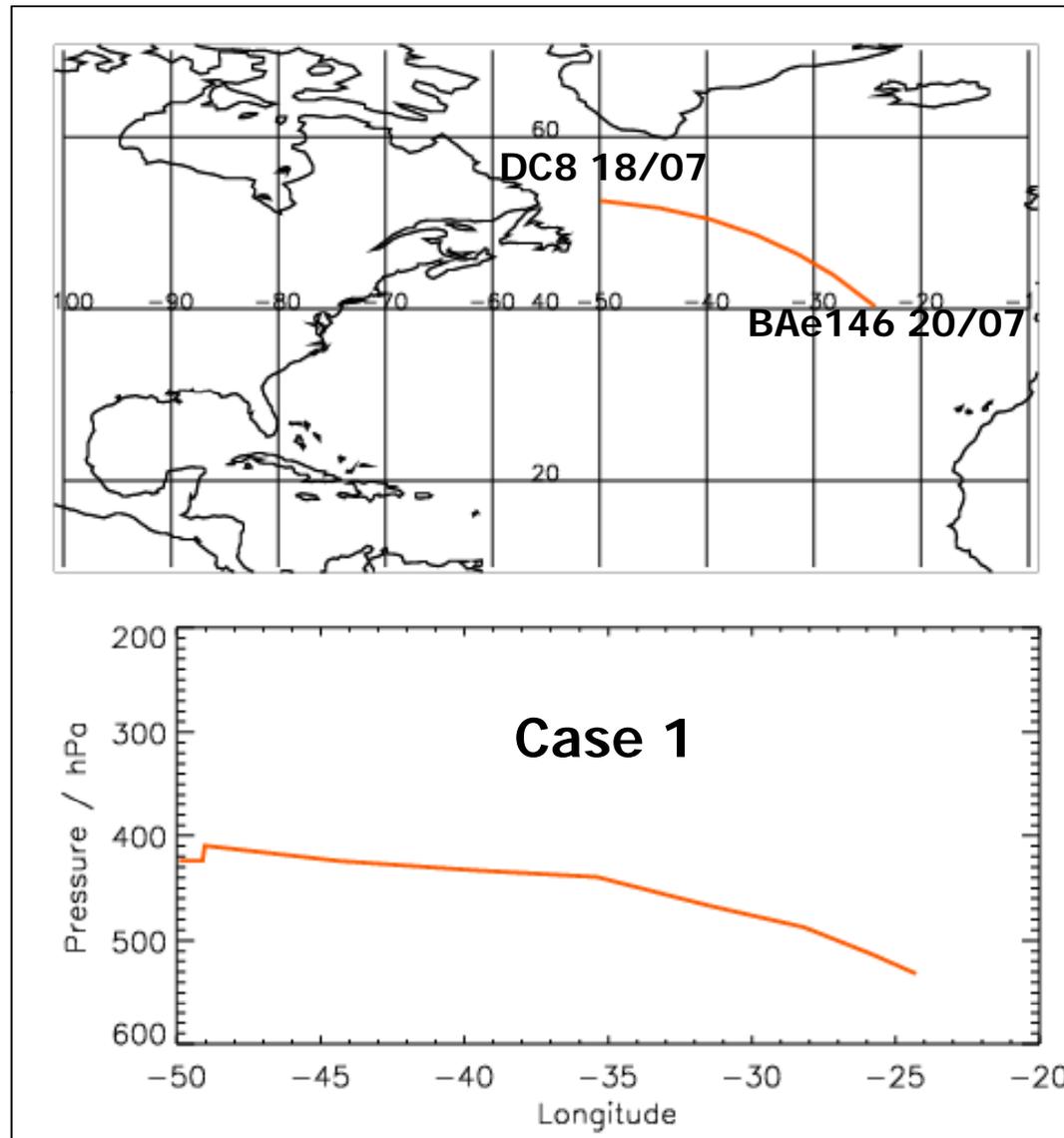


Dots: simulations
Lines: observations

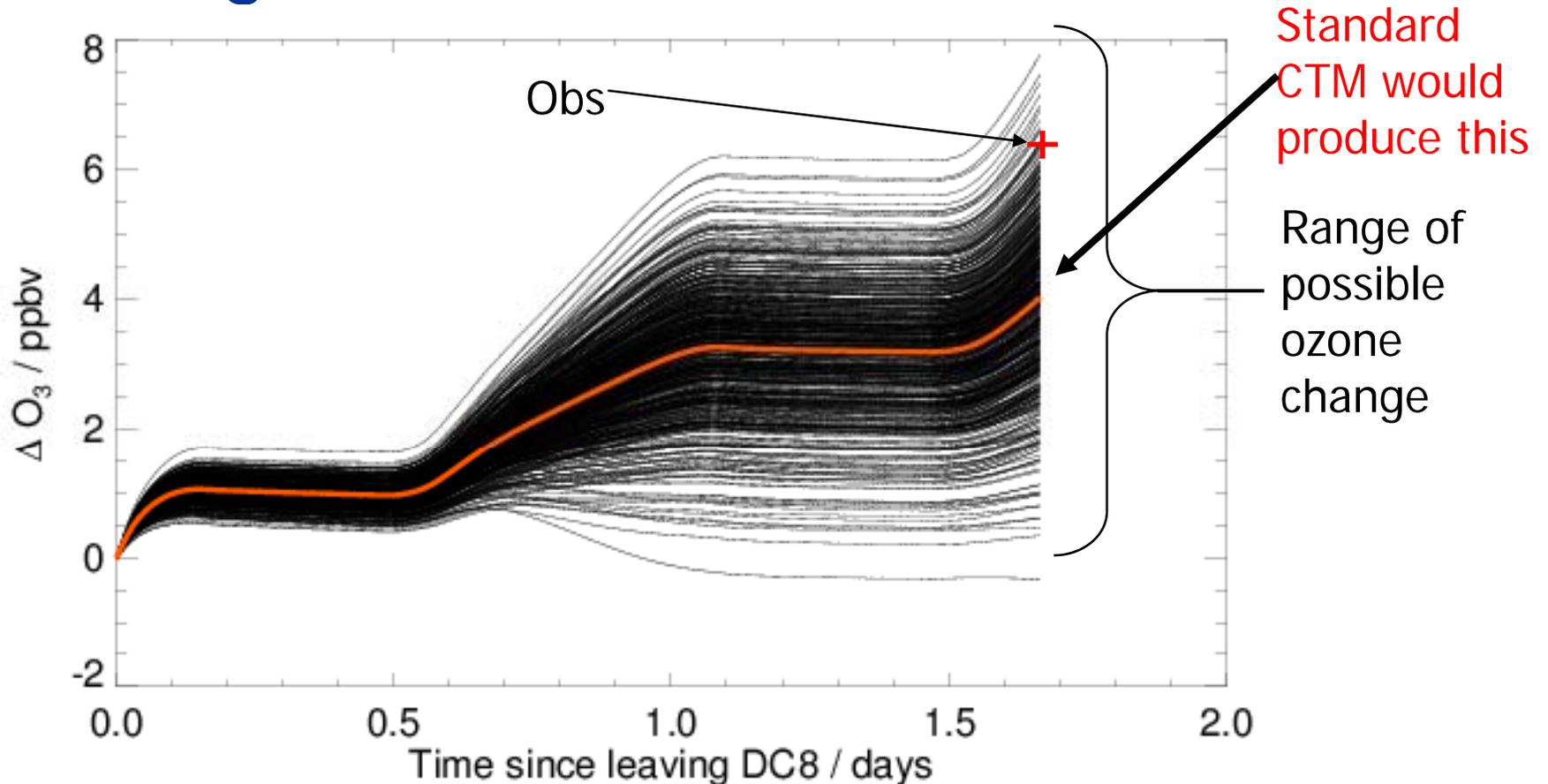
DC8 – 18/07
Bae-146 – 20/07
Falcon – 23/07

Some conclusions from the Real et al., ACPD, 2008 paper:

- The Lagrangian simulation reproduces the observed mean concentrations
- The evolution of O_3 is dominated by chemical phenomena versus mixing phenomena for CO
- Net O_3 production during transport of about 4 ppbv/day - 80 % due to PAN decomposition.
- Aerosols have a strong impact on the reduction of photochemistry (15 % of net O_3 production).
- HNO_3 concentrations are significantly depleted during transport because of wet deposition
- HNO_3 photolysis leads to a sustainable production of NO_x , and thus ozone
- This, in turn, leads to OH production (enhanced water vapor) with some implication for the evolution of the CO concentrations (- 50 ppbv in 5 days)

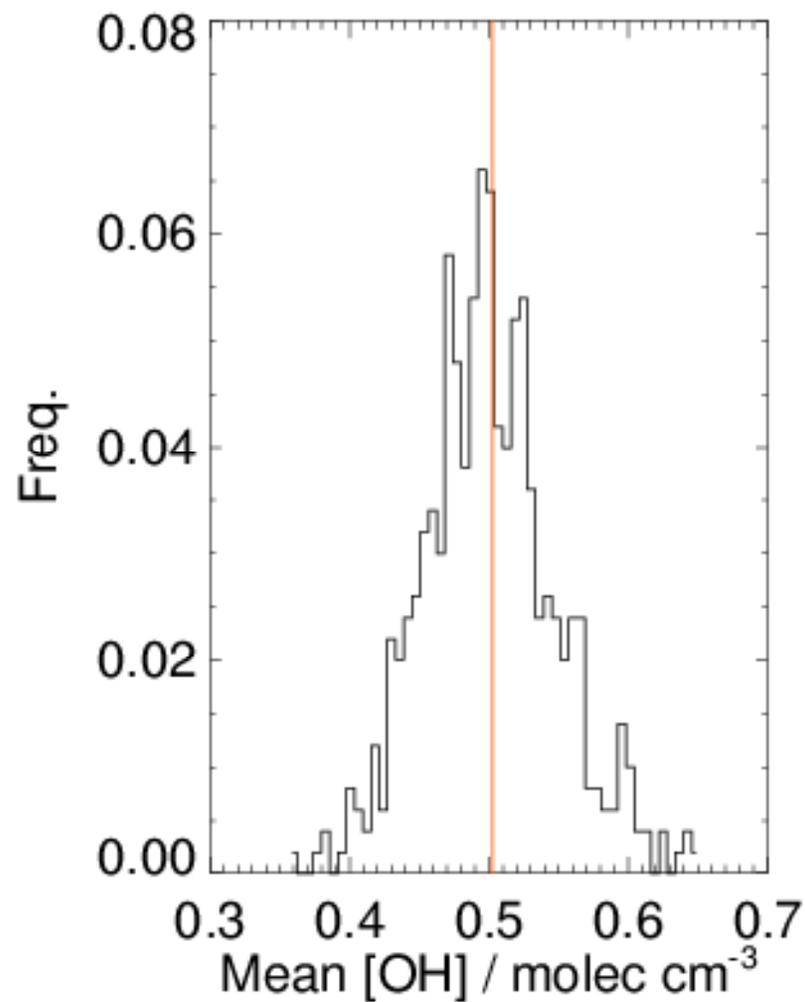
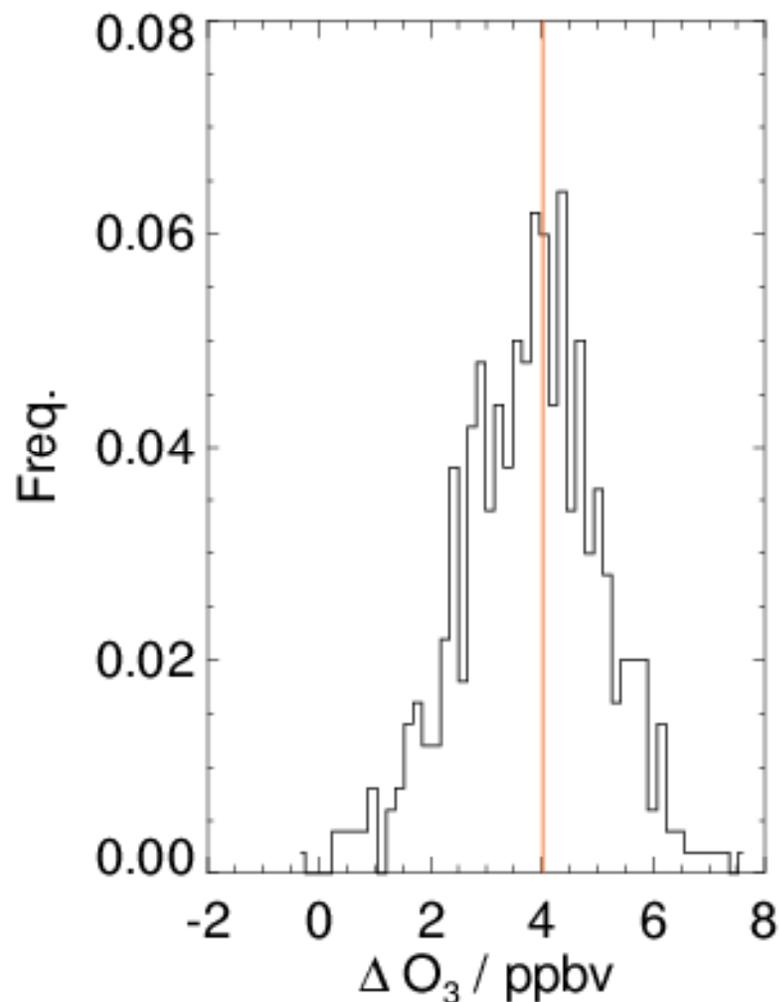


Perturbing bimolecular rate coefficients

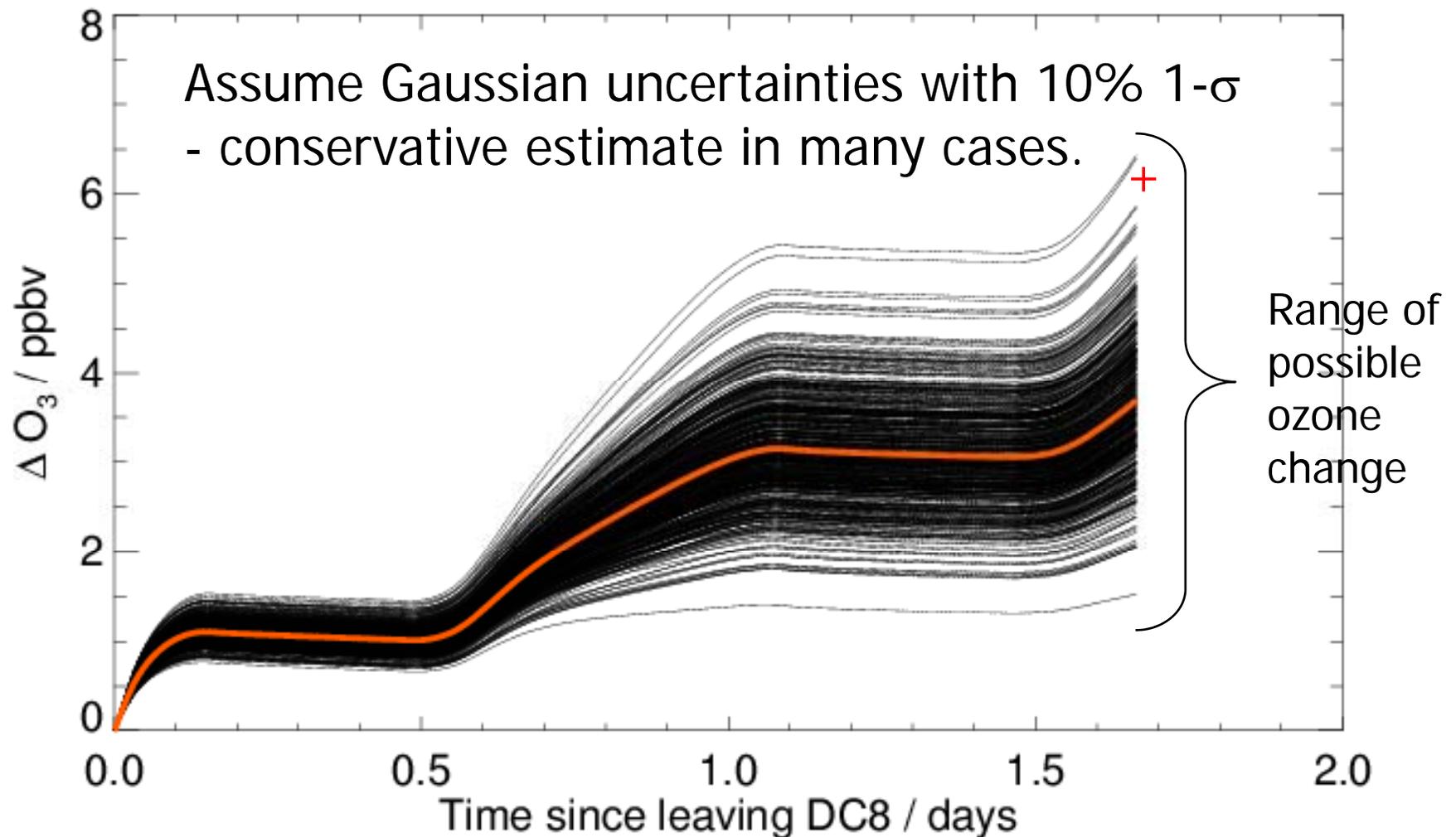


- Uncertainties from IUPAC / JPL
- Perturbed using Latin-Hypercube method

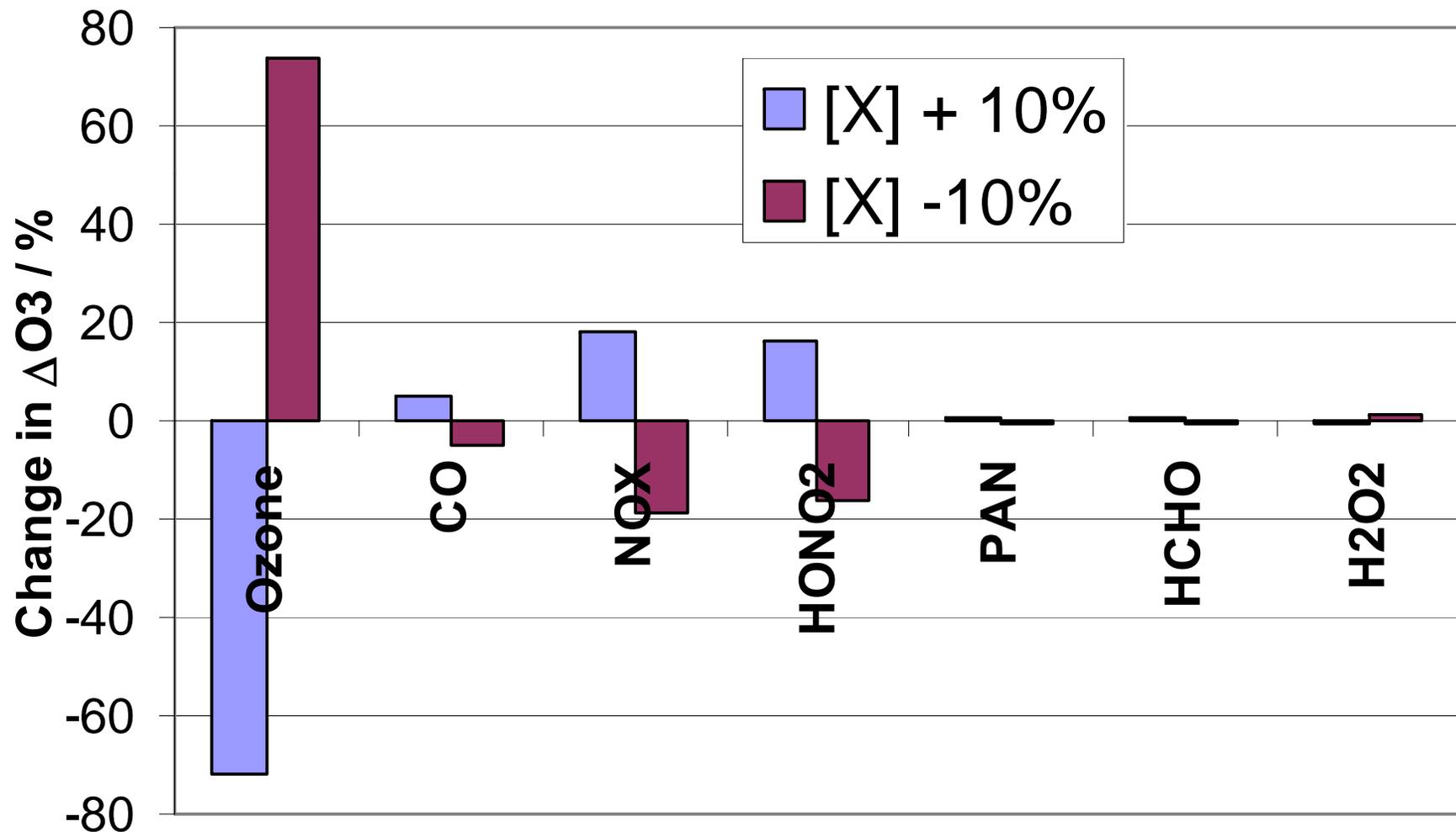
Perturbing bimolecular rate coefficients



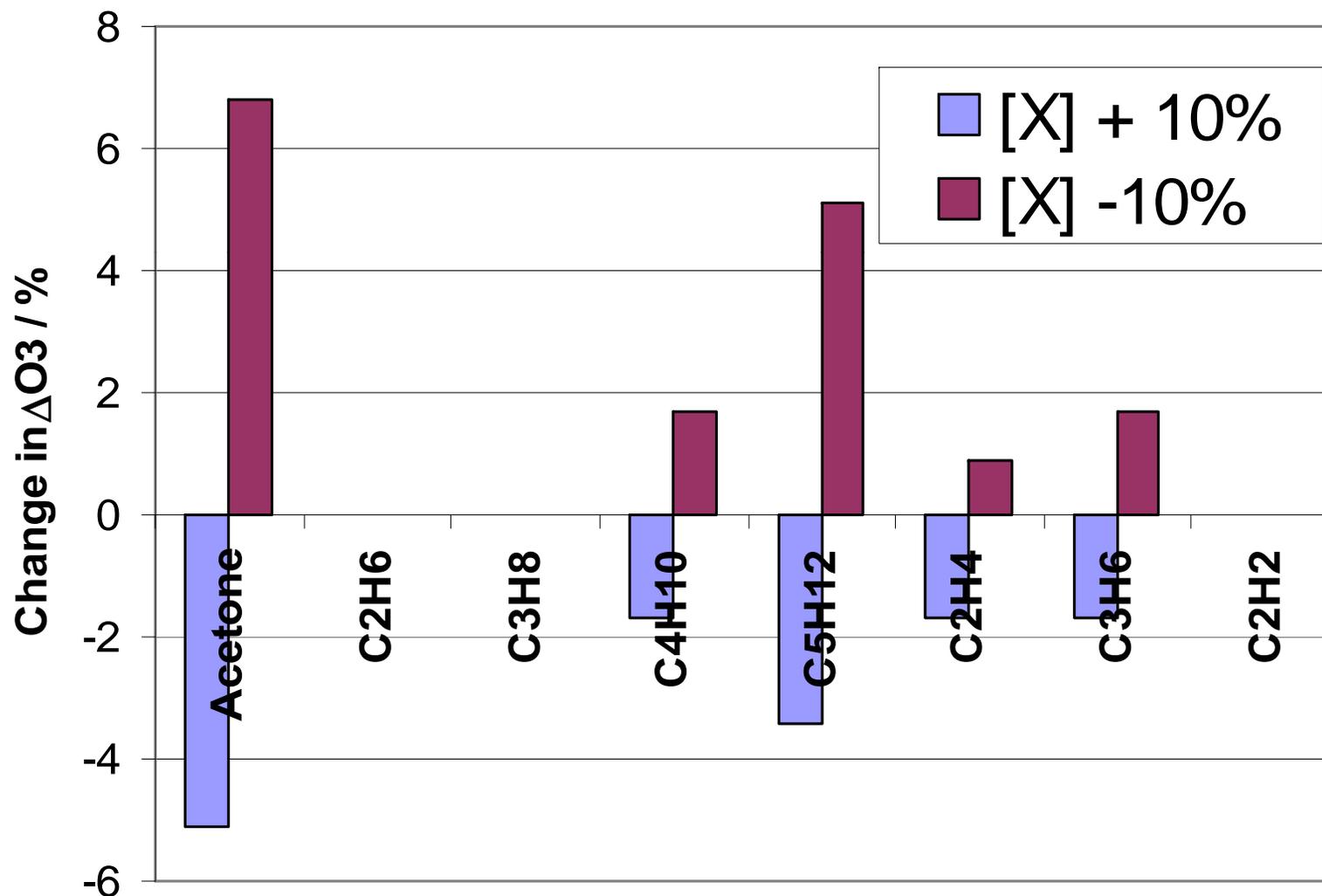
Perturbing initial concentrations



Impact on trajectory ΔO_3 for 10% perturbation to initial concs



Impact on trajectory ΔO_3 for 10% perturbation to initial concs



- **Which rate constants and other model parameters produce largest sensitivities?**
- **Which rate constants are a priority for further investigation?**
- **Which in-situ obs are key to understanding chemical evolution of different plumes (biomass, anthrop)?**
- **Which instruments are a priority for improvement?**

The HTAP Experiment Set 3 should offer
unique way of consistently comparing
observations with a wide range of models.

Very useful resource to the community

But we need to know to know when the
observations are telling us something useful
and when they are not



➔ **Time line**

- Proposal uploaded on the HTAP wiki web page end of February
- Model outputs are getting uploaded (or will be in the next weeks)
- Model outputs likely to be accepted until end of July
- First results should be available in this coming fall

➔ **Proposed analyses**

- General characterisation of air masses over the North Atlantic area
- On-route processing of plumes
 - case studies
 - “ensemble” of plumes
- Aerosol export (export efficiency of black carbon aerosols)
- Impact of injection height on of biomass burning emissions

➔ **What is next?**

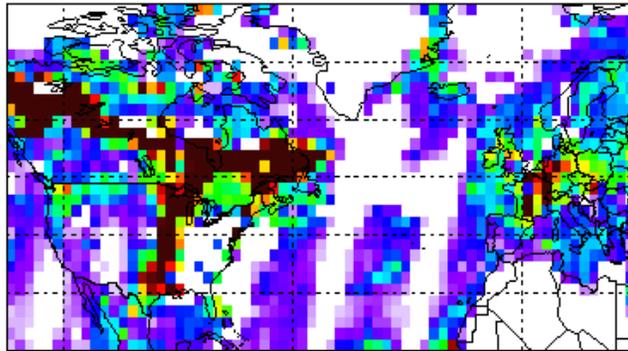
- Others analyses?
- TP simulations in support of ES?
- With this set of simulations, it will be difficult to determine why the models may differ => Try to link with ACC ? (already link to GEMS).



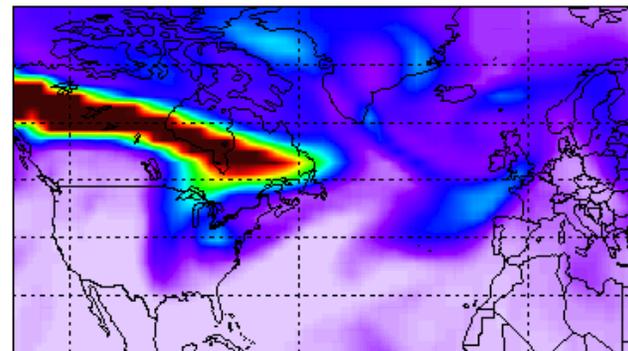
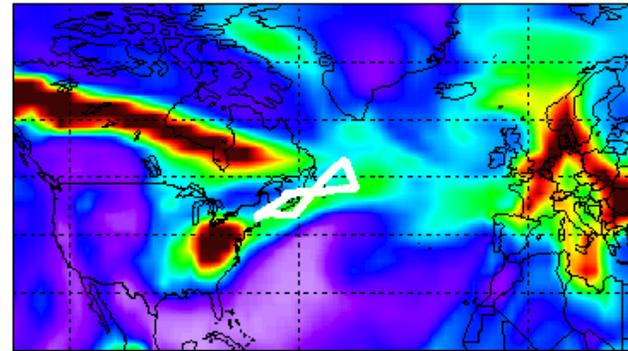
The HTAP

July 18

MODIS AOD fine mode



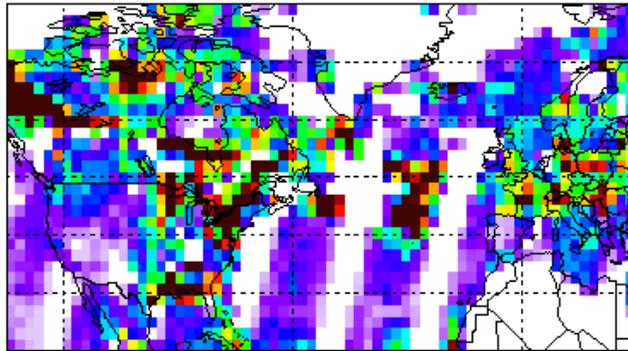
GEOS-Chem AOD fine mode



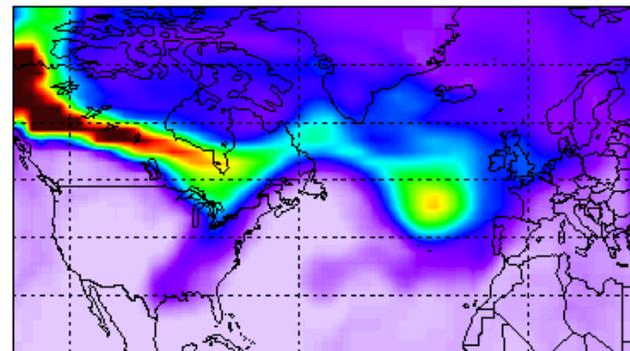
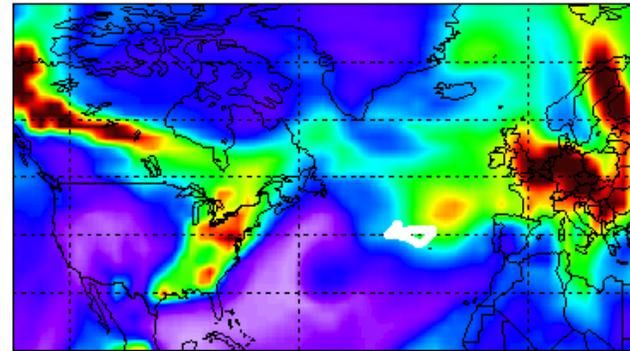
North American biomass burning AOD

July 20

MODIS AOD fine mode



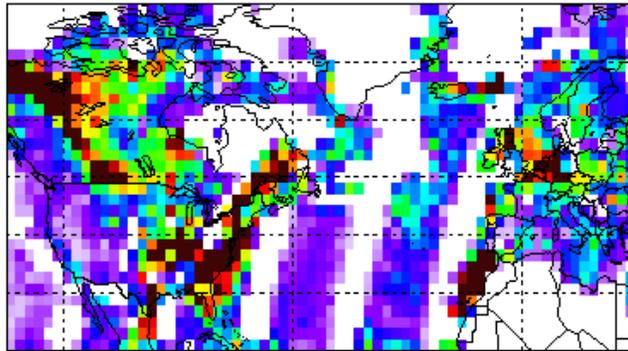
GEOS-Chem AOD fine mode



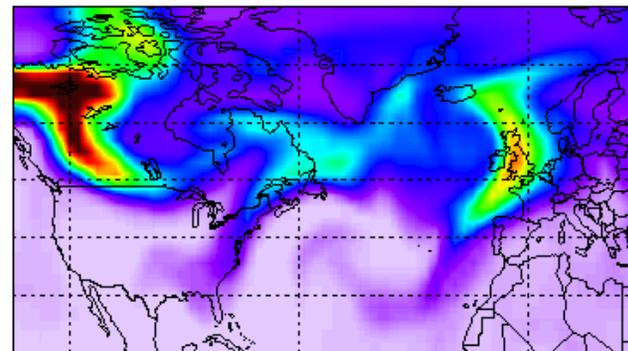
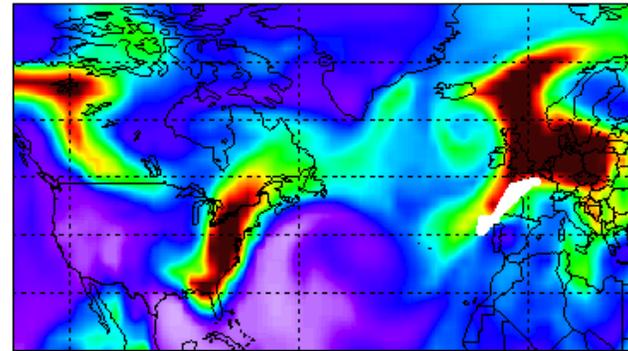
North American biomass burning AOD

July 22

MODIS AOD fine mode



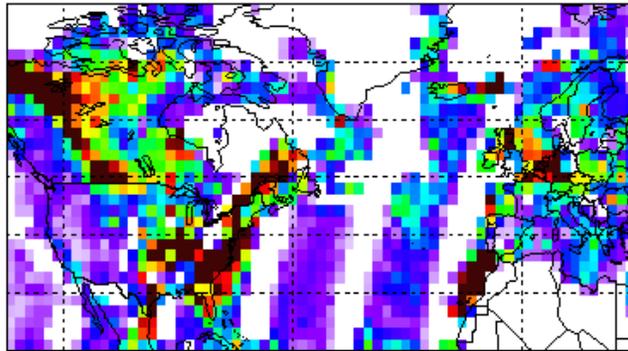
GEOS-Chem AOD fine mode



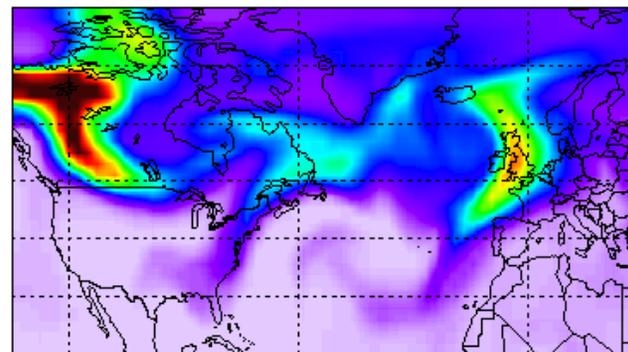
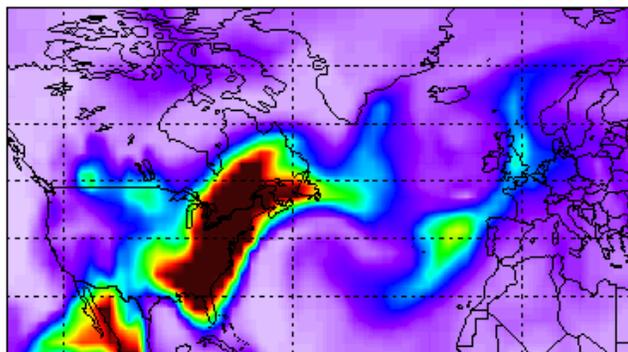
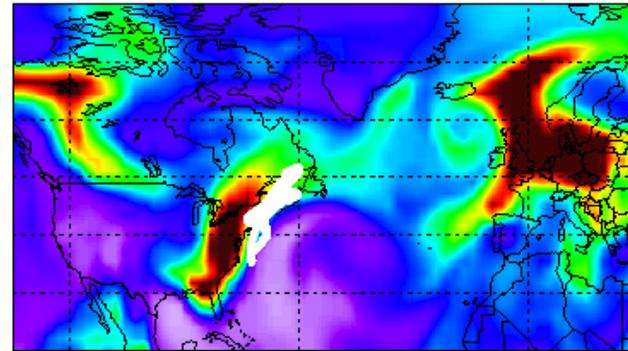
North American biomass burning AOD

July 22

MODIS AOD fine mode



GEOS-Chem AOD fine mode

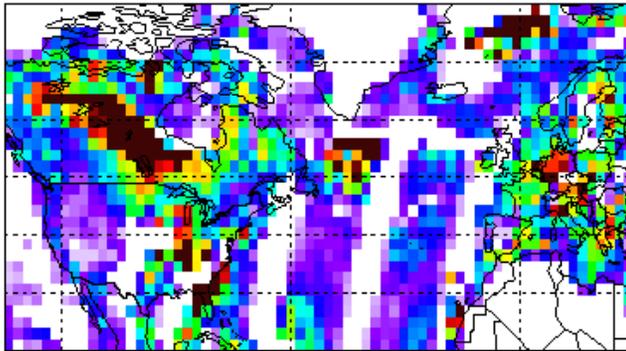


North American anthropogenic AOD

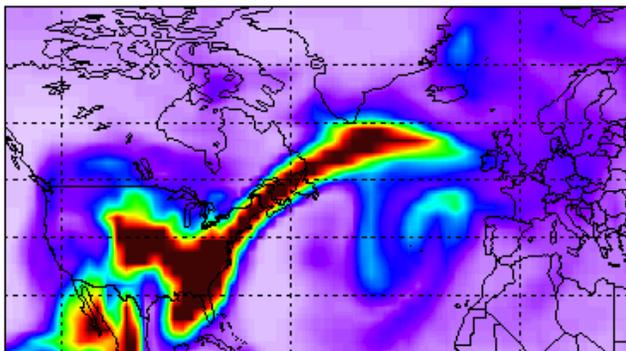
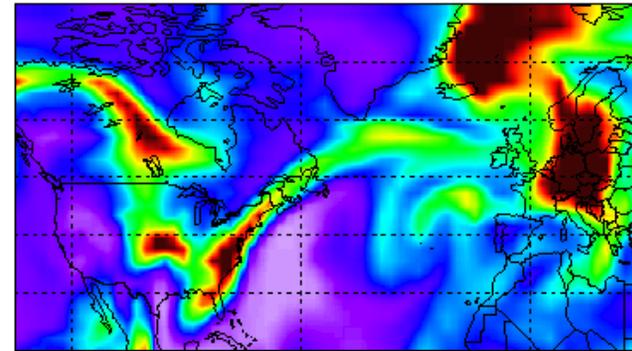
North American Biomass burning AOD

July 24

MODIS AOD fine mode



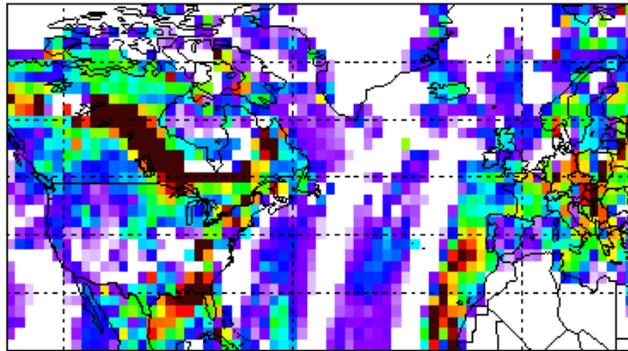
GEOS-Chem AOD fine mode



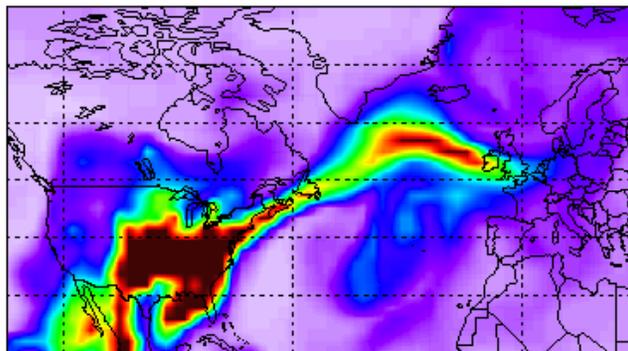
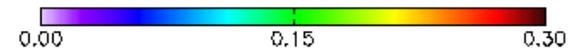
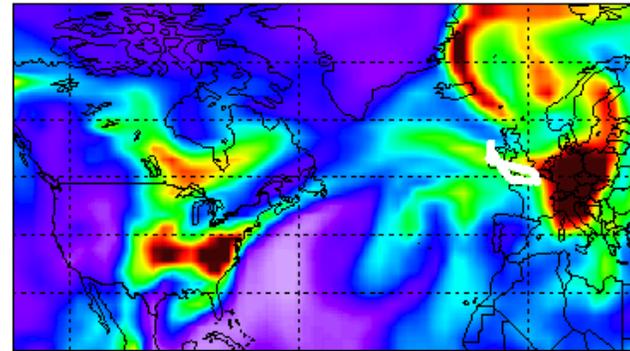
North American anthropogenic AOD

July 25

MODIS AOD fine mode



GEOS-Chem AOD fine mode



North American anthropogenic AOD



Main conclusions from the Real et al., JGR [2007a] paper:

- The Lagrangian simulation reproduces the observed mean concentration and evolution of correlations
- The evolution of O₃ is dominated by chemical phenomena versus mixing phenomena for CO
- Net O₃ production during transport of about 4 ppbv/day - 80 % due to PAN decomposition.
- Aerosols have a strong impact on the reduction of photochemistry (15 % of net O₃ production).

Some processes we can test in the model:

- Chemical evolution
- Mixing
- Influence on receptor regions

Some further considerations:

- This Alaskan anthropogenic plume is a “good candidate” to examine intercontinental transport of O₃ because of the different processes occurring in route
- Transport of both ozone pollution and aerosols can be addressed
- This plume significantly affects Europe, both in terms of ozone and aerosols
- “Biomass burning” pollution : Is that still relevant for HTAP?

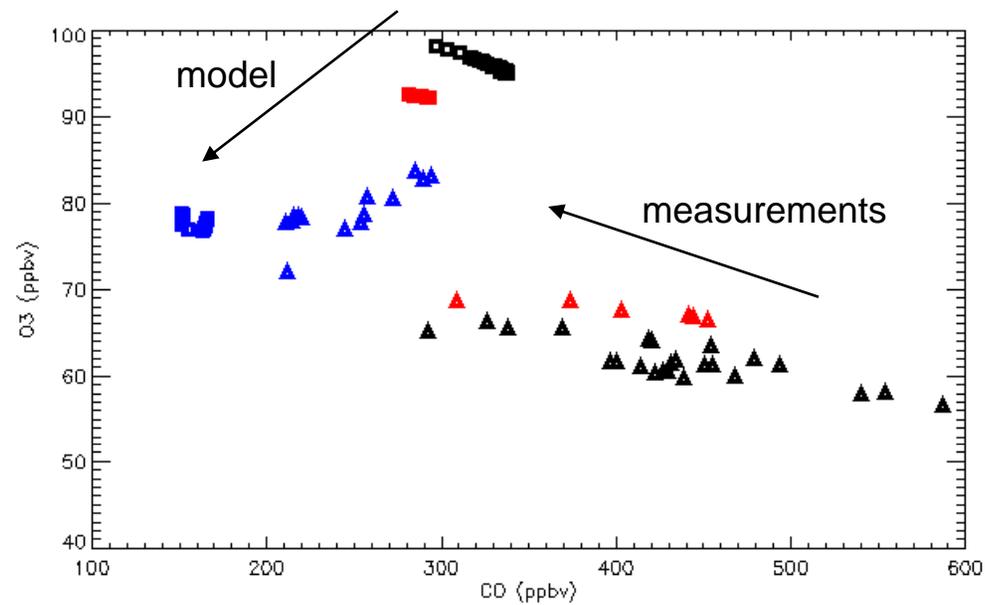


Main conclusions from the Real et al., JGR [2007b] paper:

- The Lagrangian simulation reproduces the observed mean concentrations
- HNO_3 concentrations are significantly depleted during transport because of wet deposition
- HNO_3 photolysis leads to a sustainable production of NO_x , and thus ozone, which, in turn, leads to OH production (enhanced water vapor) with some implication for the evolution of the CO concentrations (-50 ppbv in 5 days)

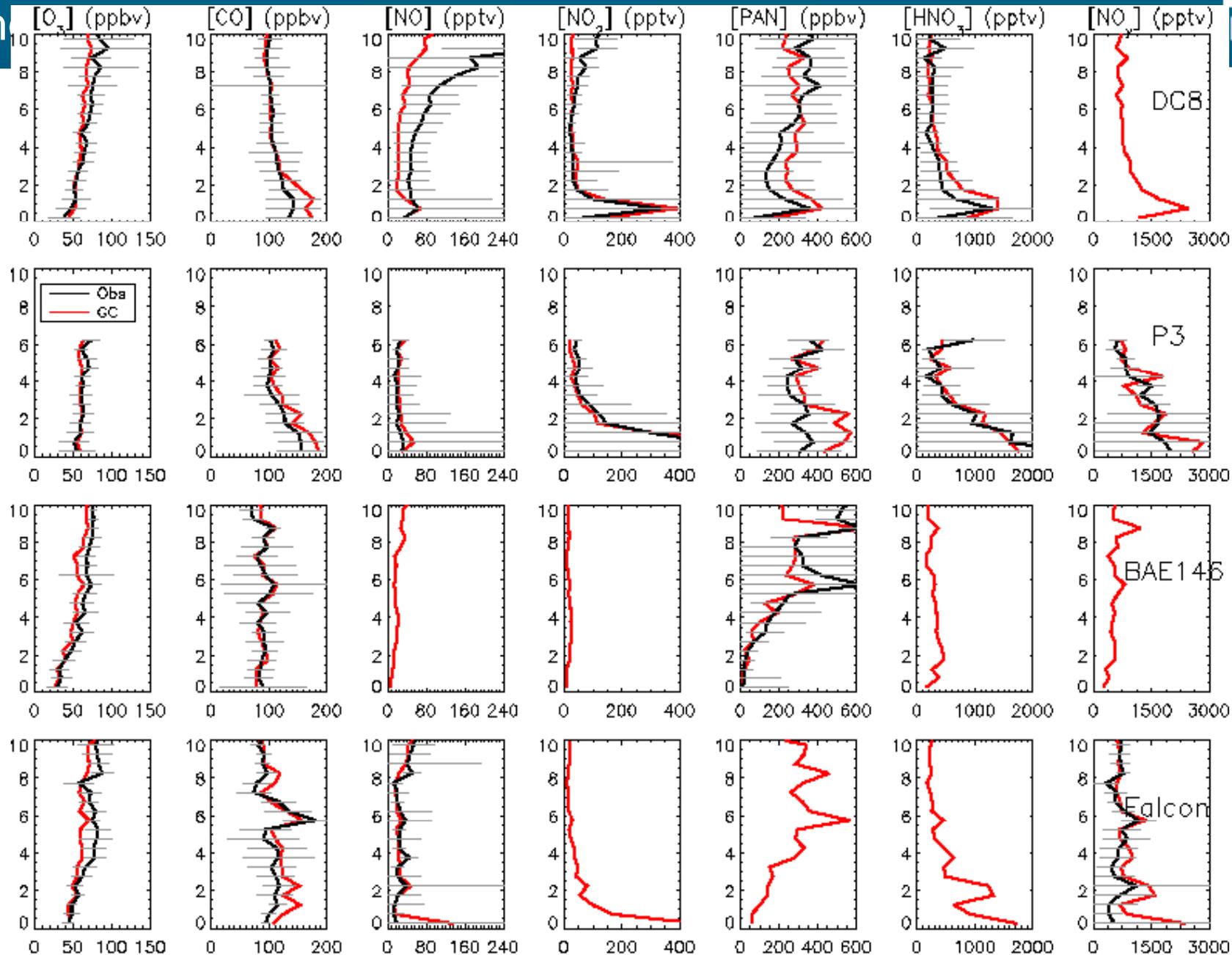
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- Chemical evolution
- Wet deposition
- Mixing





Sch





Sch

