Tropospheric Airborne Measurement Evaluation Panel

The HTAP Perspective and beyond

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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]
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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Mat Evans + Isabelle Bey
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

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

Upper Tropo

Upper Outflow

Mid Troposphere

Low Outflow

Marine

Biomass burning

Pressure (hPa)
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. Bousserez and J.-L- Attié, Laboratoire d’aérologie, Toulouse, France.
Within each cluster are the relationships species the same?
Principal components analysis will allow us to investigate this

**observations**

- "clean" lower trop.
- Biomass burning influenced air masses
- Middle-upper troposphere
- "polluted" lower trop.

**model**
• 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

Isabelle Bey + Mat Evans
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

Auvray et al., JGR, 2007
GEOS-Chem 02/04/1997

Mean sea level pressure (hPa)

H₂O (ppmv) at 700 hPa

ECHAM5-MOZ

Auvray et al., JGR, 2007
Impact of North American outflow over the North Atlantic – April 1997
• 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)
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Rojkin Park
\[ f_X(z) = \frac{1}{R_X} \left( \frac{\Delta[X]}{\Delta[CO]} \right)(z) \]

\[ f_{X_{\text{norm}}}(z) = \frac{f_X(z)}{f_X(0)} \]

- \( X = \) combustion-derived species
- \( R_X = \) emission ratio (X/CO)
- \( \Delta = \) enhancements relative to background


[Koike et al., 2003; Parrish et al, 2004]
timescale $\tau$ for conversion of hydrophobic to hydrophilic BC in global models
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Solene Turquety
• General characterisation of chemical signatures of different air masses over the North Atlantic area
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Kathy Law + Steve Arnold
Real et al., 2008
On-route processing of plumes of biomass burning and anthropogenic origins: anthropogenic plume case study

Real et al., ACPD, 2008
On-route processing of plumes of biomass burning and anthropogenic origins: case study of a biomass burning plume

Dots: simulations
Lines: observations

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

Real et al., JGR, 2007
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 NOx, 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
Assume Gaussian uncertainties with 10% 1-σ - conservative estimate in many cases.

Range of possible ozone change
Impact on trajectory $\Delta O_3$ for 10% perturbation to initial concs

- **Ozone**: Large increase
- **CO**: Small increase
- **NOx**: Small increase
- **HONO**: Moderate increase
- **PAN**: Negligible change
- **HCHO**: Negligible change
- **H2O2**: Negligible change

Legend:
- [X] + 10%
- [X] -10%
Impact on trajectory $\Delta O_3$ for 10% perturbation to initial concs

- Acetone: $\Delta O_3 +10\%$
- $[X] + 10\%$
- $[X] -10\%$

Graph showing changes in $\Delta O_3$ for various compounds.
• 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 a unique way of consistently comparing observations with a wide range of models.

Very useful resource to the community

But we need to know when the observations are telling us something useful and when they are not.
HTAP Experiment Set 3 – time line, 2

**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
Long range transport events observed during ICARTT

July 18

MODIS AOD fine mode

GEOS-Chem AOD fine mode

North American biomass burning AOD
Long range transport events observed during ICARTT

July 20

MODIS AOD fine mode

GEOS-Chem AOD fine mode

North American biomass burning AOD
Long range transport events observed during ICARTT

July 22

MODIS AOD fine mode

GEOS-Chem AOD fine mode

North American biomass burning AOD
Long range transport events observed during ICARTT

July 22

MODIS AOD fine mode

GEOS-Chem AOD fine mode

North American anthropogenic AOD

North American Biomass burning AOD
Long range transport events observed during ICARTT

July 24

MODIS AOD fine mode

GEOS-Chem AOD fine mode

North American anthropogenic AOD
Long range transport events observed during ICARTT

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 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 decompositon.
- Aerosols have a strong impact on the reduction of photochemistry (15 % of net O$_3$ 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 O$_3$ 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 NOx, 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)

Some processes we can test in the model:
- Chemical evolution
- Wet deposition
- Mixing
1.1. Analysis of a specific event of biomass burning pollution transport

measurements

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

model

measurements

MOCAGE
The ICARTT campaign (summer 2004)
The ICARTT campaign (summer 2004)