

# Intercontinental Transport of Ozone and Precursors (ITOP)

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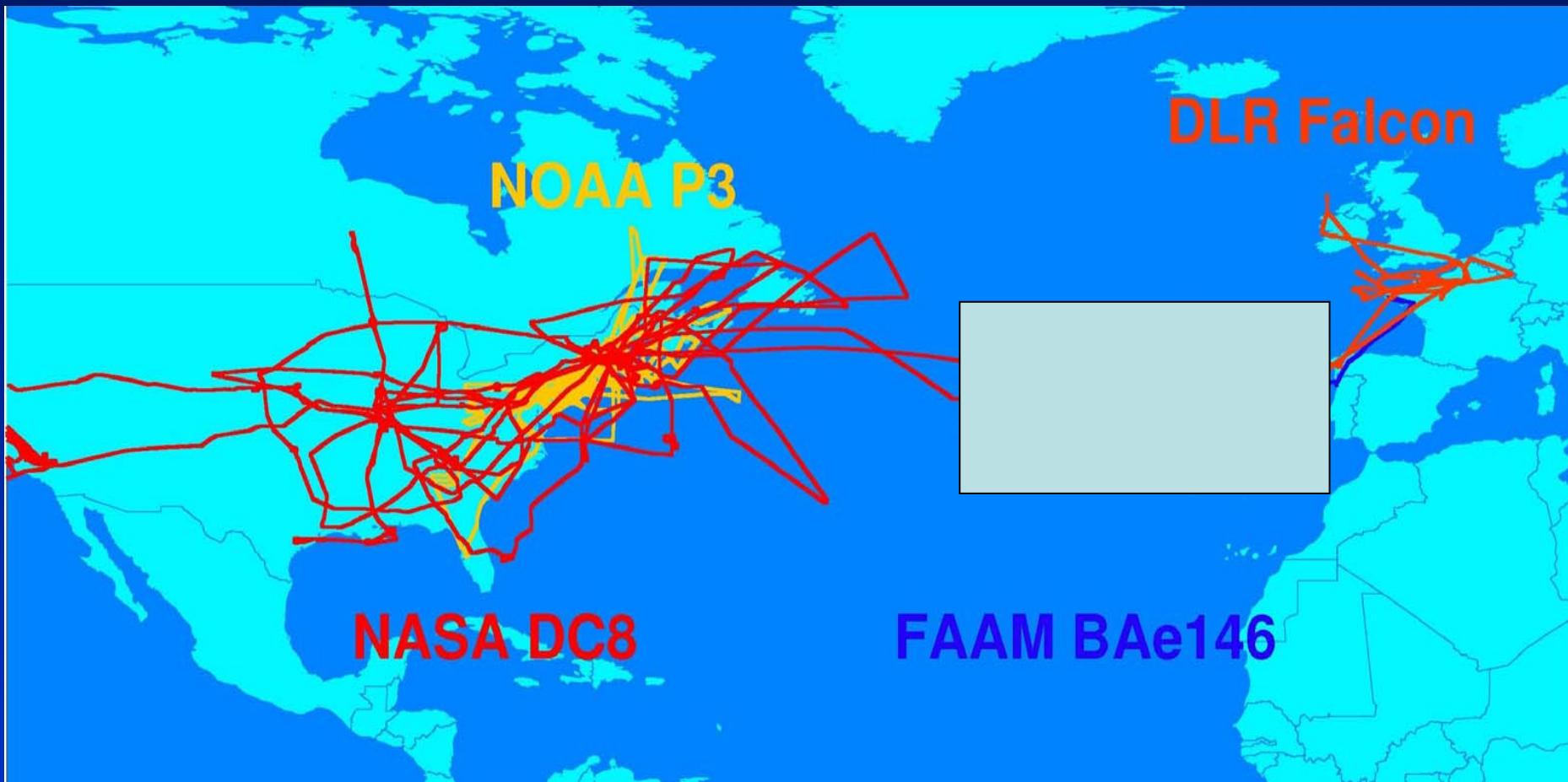
*DirectFlight Ltd*

Martin Darling, Andrew 'Rodders' Boardman, Simon Tooley

*Avalon Aero Ltd*

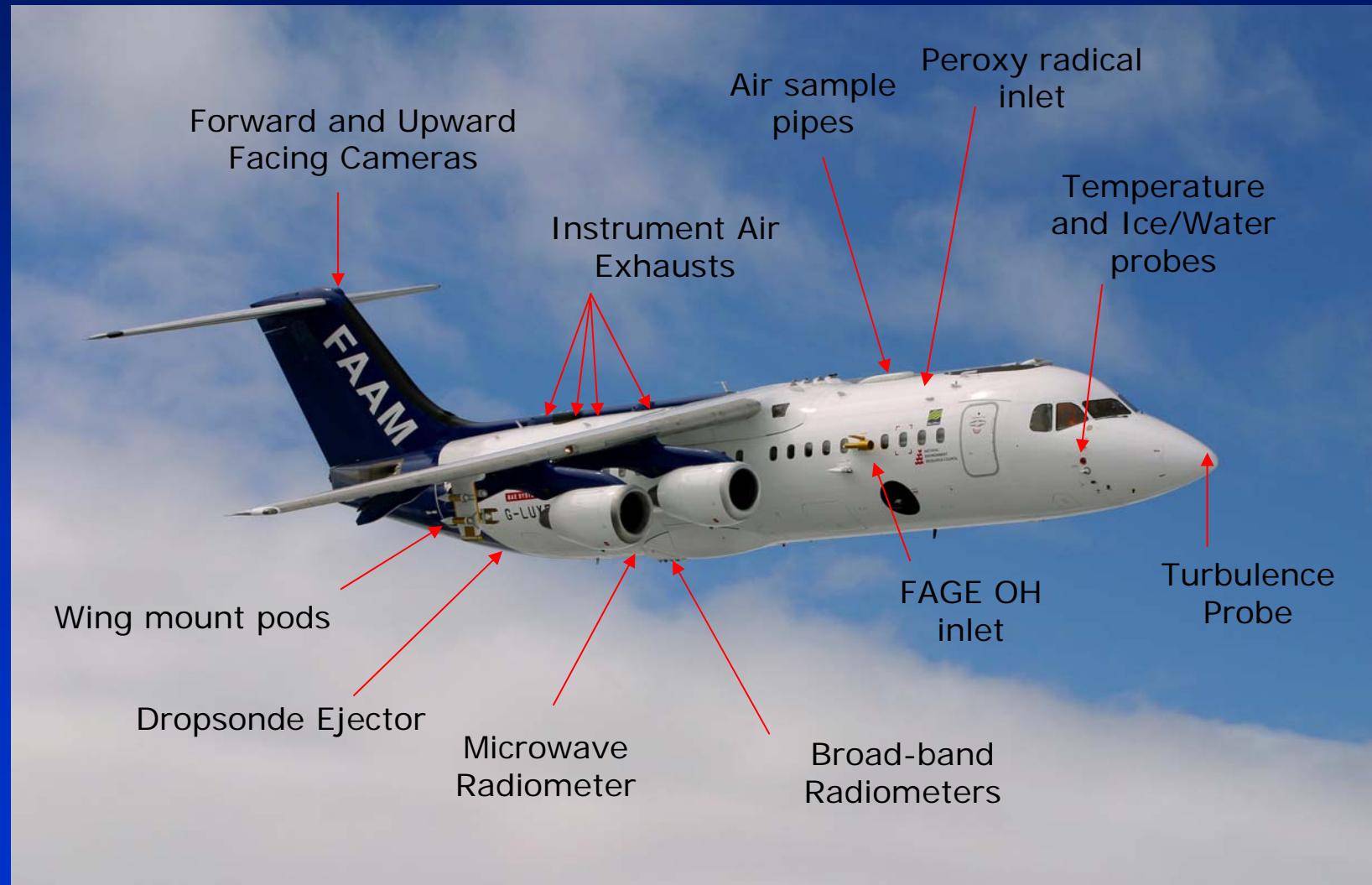


# ICARRT Summer 2004



*FAAM BAe146, based in Faial, Azores, flew 12 science flights ~70 hours including intercomparison flights with DLR Falcon and NASA DC8*

# The Facility for Airborne Atmospheric Measurements BAe-146 Research Aircraft



# Flight Highlights

- Three-point Lagrangian opportunities: 19/7 + 25/7  
28/7 + 29/7 and 31/7 + 1/8.
- Two-point Lagrangian opportunities: 17/7, 20/7 (fire).
- Forest fires: 15/7, 19/7, 20/7, 31/7 (with strat), 1/8.
- Pico flypass: 15/7, 17/7, 19/7, 31/7, 1/8.
- Comparisons: 28/7 (DC8), 3/8 (Falcon).
- ENVISAT underpass: 22/7.

**B028 – 12/7/04 – Fire plumes in SW approaches**

**B029 – 15/7/04 – W to E Low level pollution + forest fires**

**B030 – 17/7/04 – First Lagrangian opportunity, - skimmed P3 air**

**B031 – 19/7/04 – New York plume + fire layers**

**B032 – 20/7/04 – Dominated by Alaskan fires, low T**

**B033 – 22/7/04 – ENVISAT underpass / cyclone**

**B034 – 25/7/04 – Re interception of NY plume + African outflow**

**B035 – 28/7/04 – DC8 comparison / air exported by wcb**

**B036 – 29/7/04 – Upper level export in wcb from US + fires higher T**

**B037 – 31/7/04 – low level wcb sampled by P3, + fires + strat**

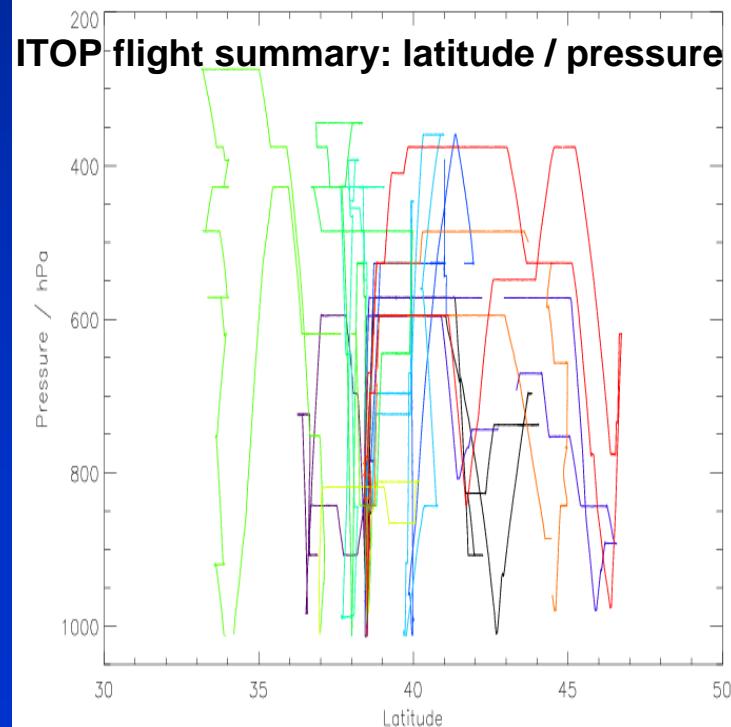
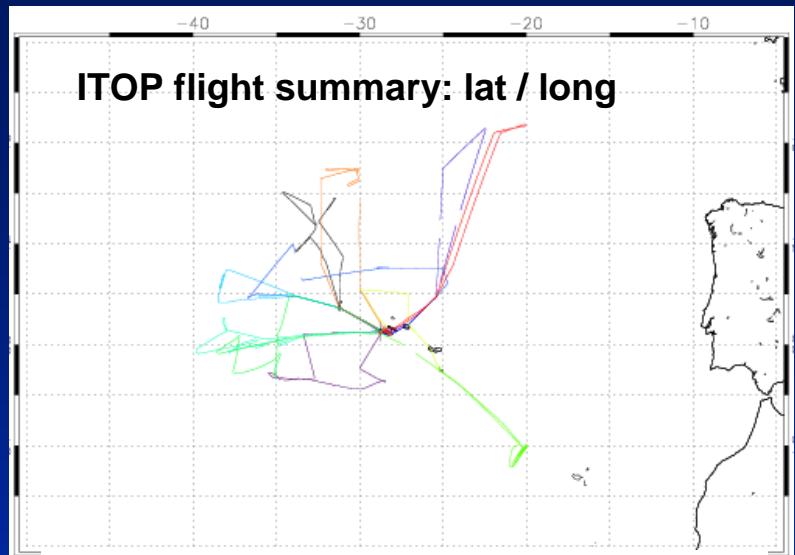
**B038 – 01/8/04 – low level wcb 2**

**B039 – 03/8/04 – Transit / Falcon intercomparison**

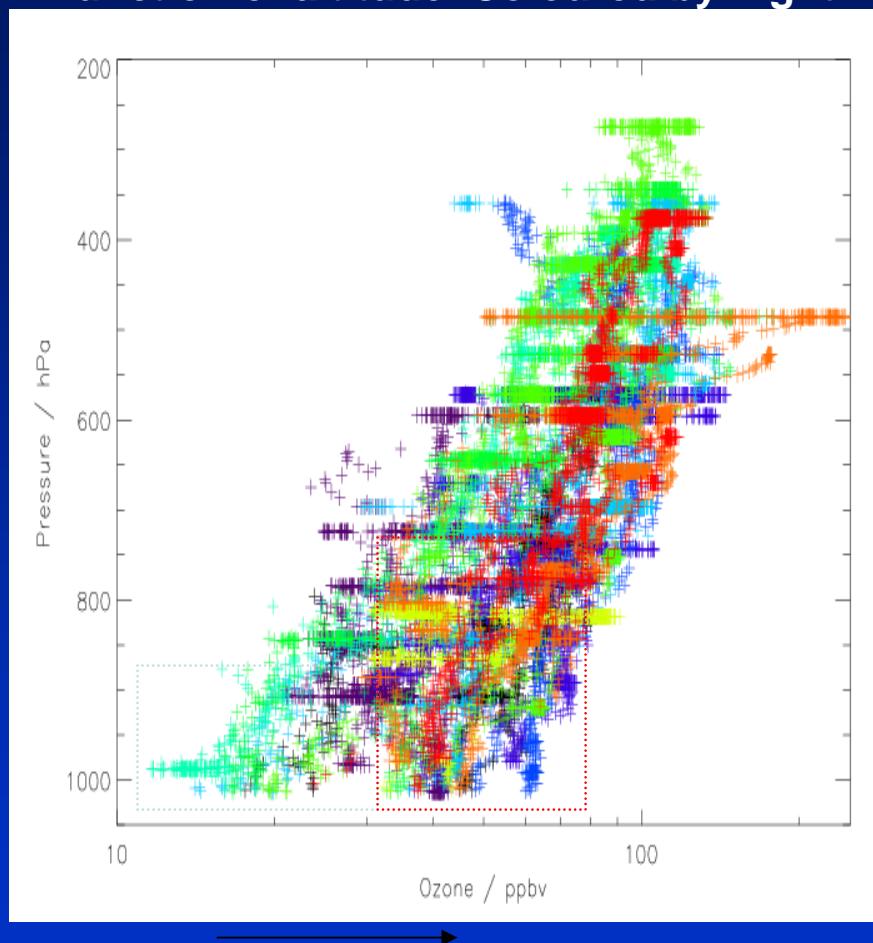
# ITOP data coverage

	B028	B029	B030	B031	B032	B033	B034	B035	B036	B037	B038	B039
Core data	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
CO	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
O <sub>3</sub>	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
NO	N	N	N	Y	Y	Y	Y	Y	Y	Y	Y	Y
NO <sub>2</sub>	N	N	N	?	?	?	?	?	?	?	?	?
NOy	?	?	?	?	?	?	?	?	?	?	?	?
HCHO	N	Y	Y	Y	Y	Y	Y	Y	Y	Y	N	N
H <sub>2</sub> O <sub>2</sub>	N	N	Y	Y	N	Y	Y	N	Y	Y	N	N
PTR-MS	N	N	N	N	N	N	N	Y	Y	N	N	N
alkyl nitrates	N	N	N	Y	Y	Y	Y	Y	Y	Y	Y	Y
Halocarbons	N	N	N	N	N	N	N	N	N	N	N	Y
PAN	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
WAS VOCs	26	60	58	61	48	52	59	32	48	61	62	34
OH/HO <sub>2</sub>	N	N	N	N	N	N	N	N	N	N	N	N
radiometers	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
PERCA	N	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
AMS	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y	Y
CPC	N	N	Y	Y	Y	Y	Y	N	Y	Y	Y	Y

# Overview of U.K. activities



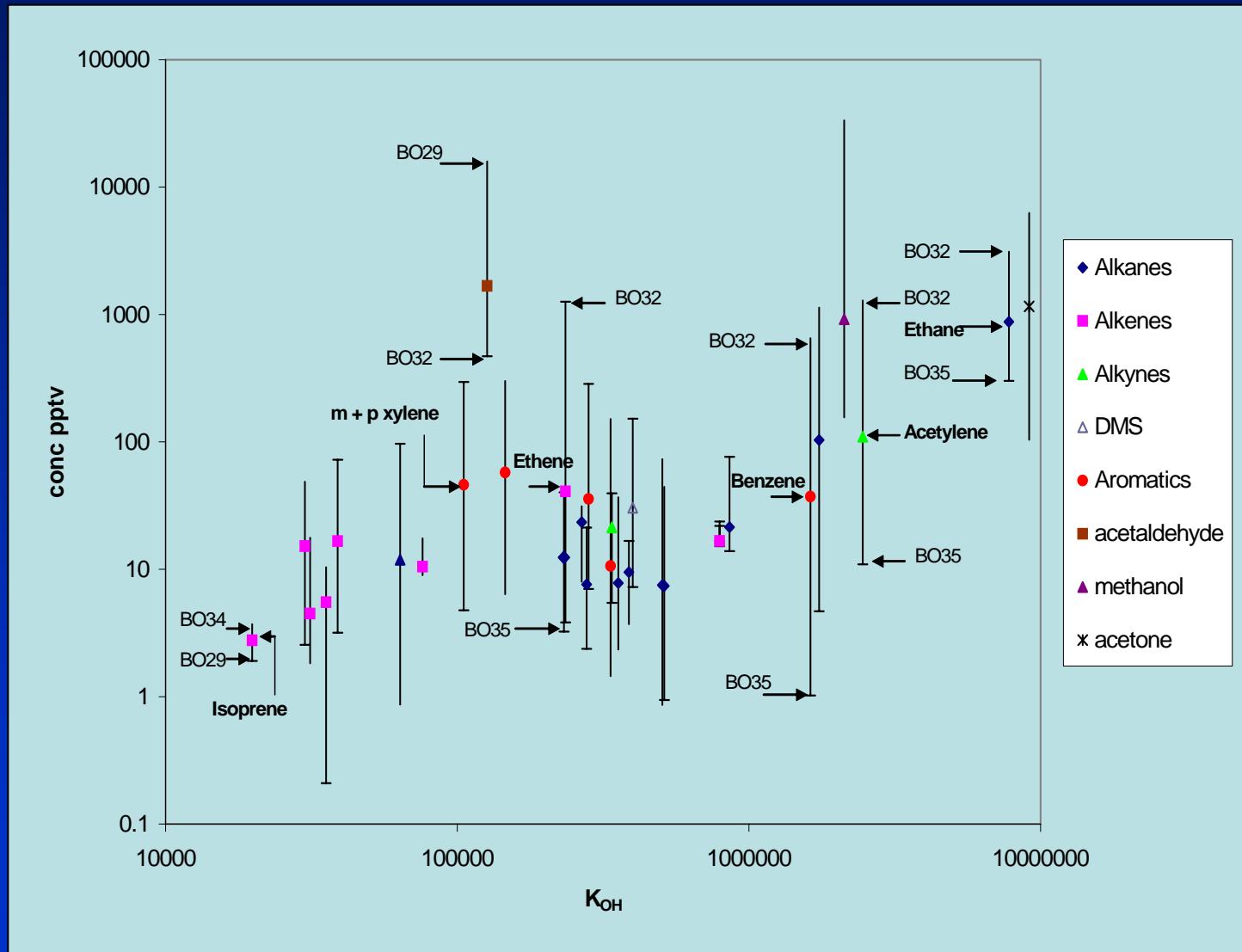
Complete ITOP campaign ozone as a function of altitude. Coloured by flight.



Clean MBL / 15 ppb

Significant long range continental influence at both low and high altitudes during campaign

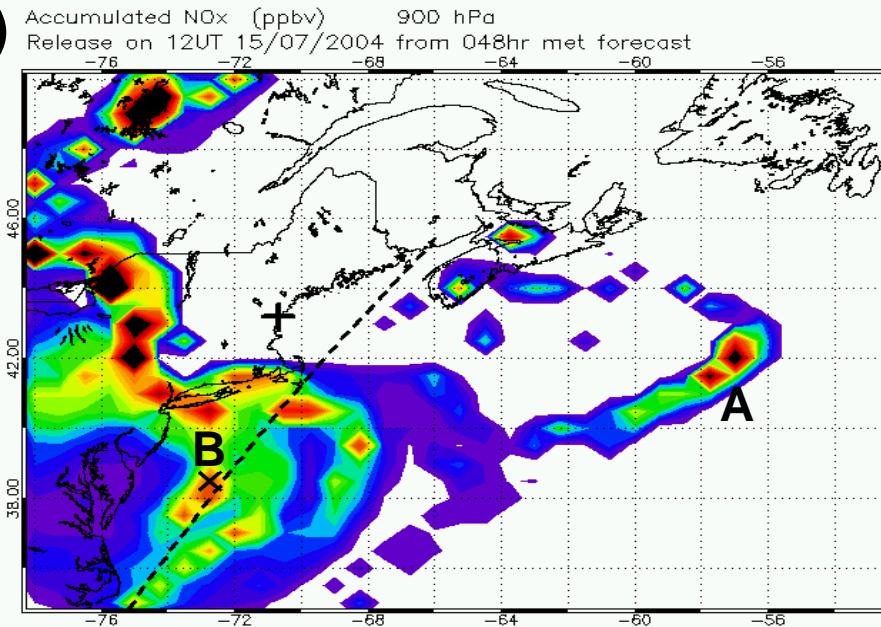
# NMHC variability – campaign overview



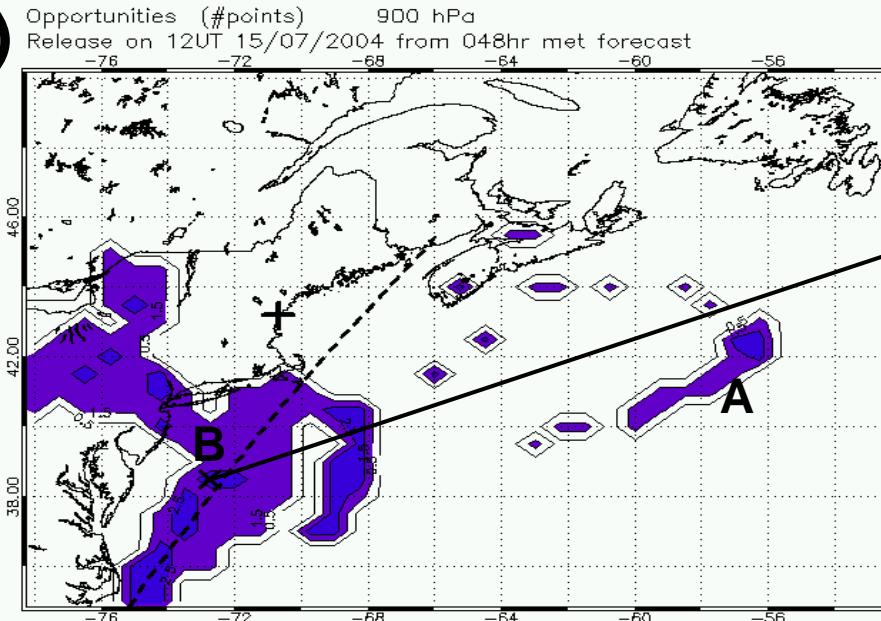
B029 / 32 fire dominated, B035 DC8 comparison

# Forecasting target for upstream domain

a)



b)

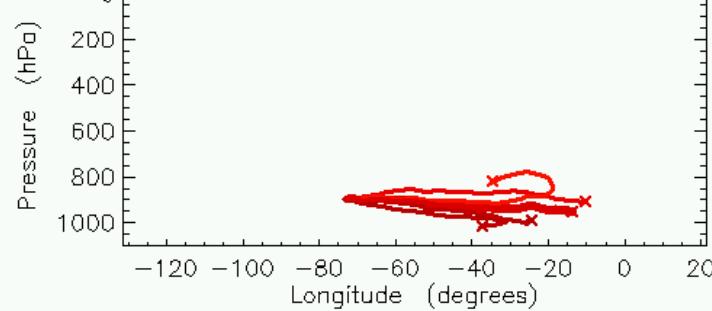
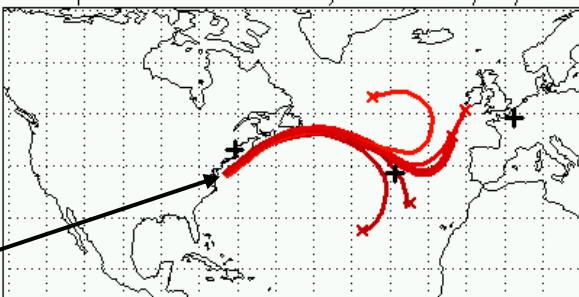


Domain filling trajectory forecasts for E.Coast America domain on 15 July.

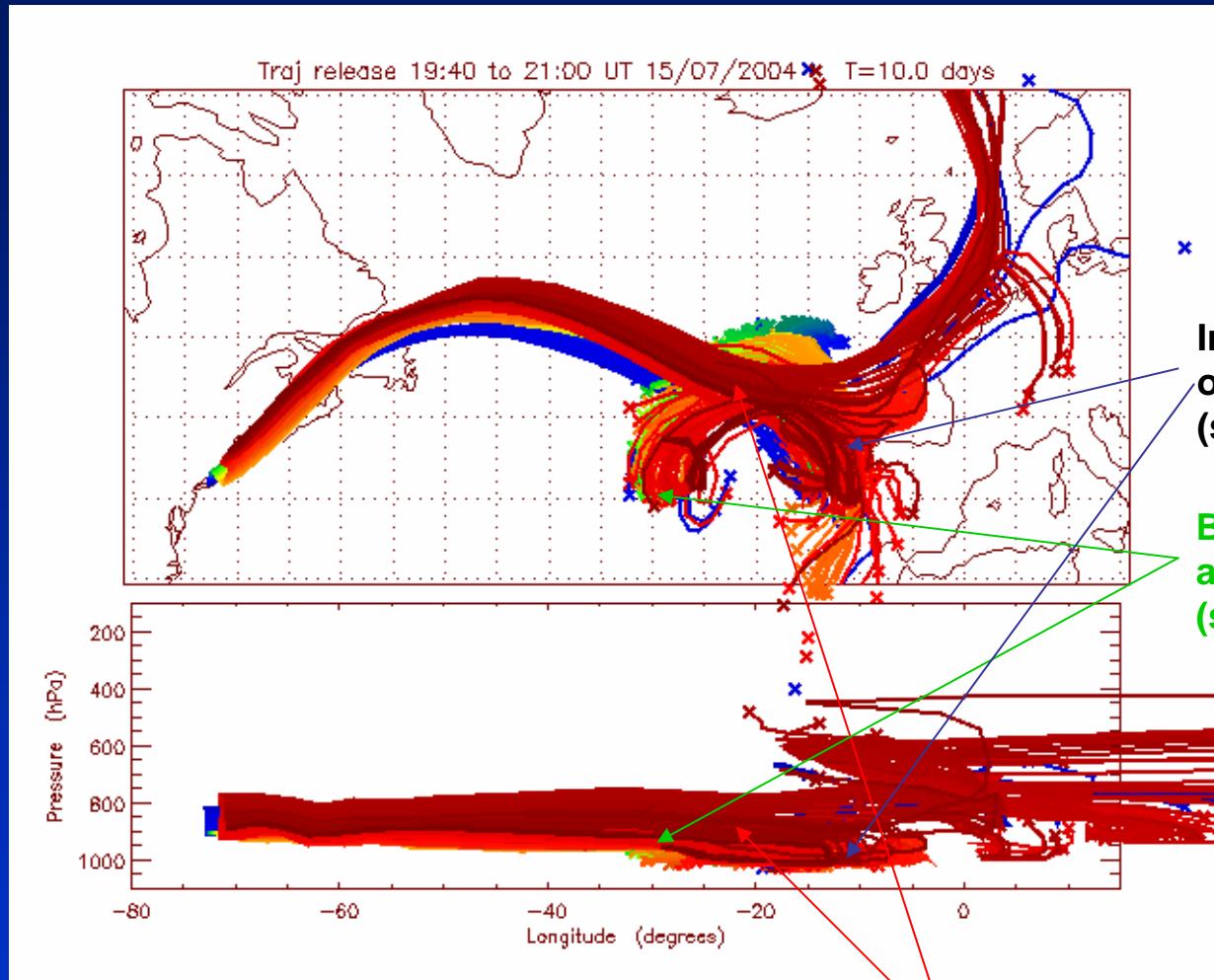
- a) NO<sub>x</sub> emissions tracer at 900 hPa
- b) 2-point and 3-point opportunities highlight targets A and B.
- c) 7 days forward from target B

$\lambda = -72$ ,  $\phi = 38.5$ ,  $p = 900 \text{ hPa}$ ,  $T = 7.00 \text{ days}$ , 12UT 15/07/2004, F+048hr

c)



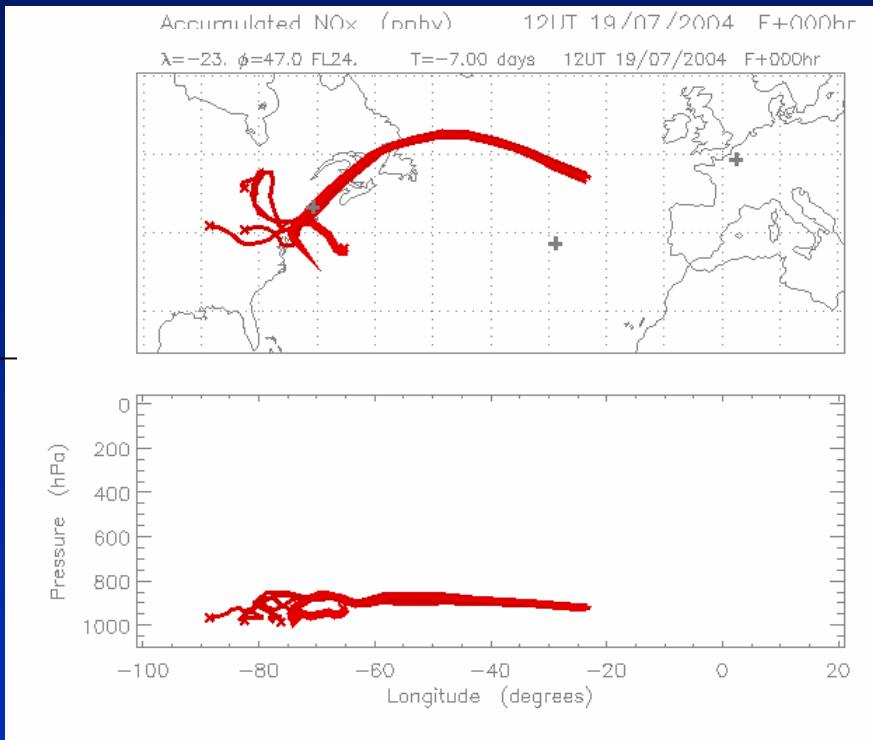
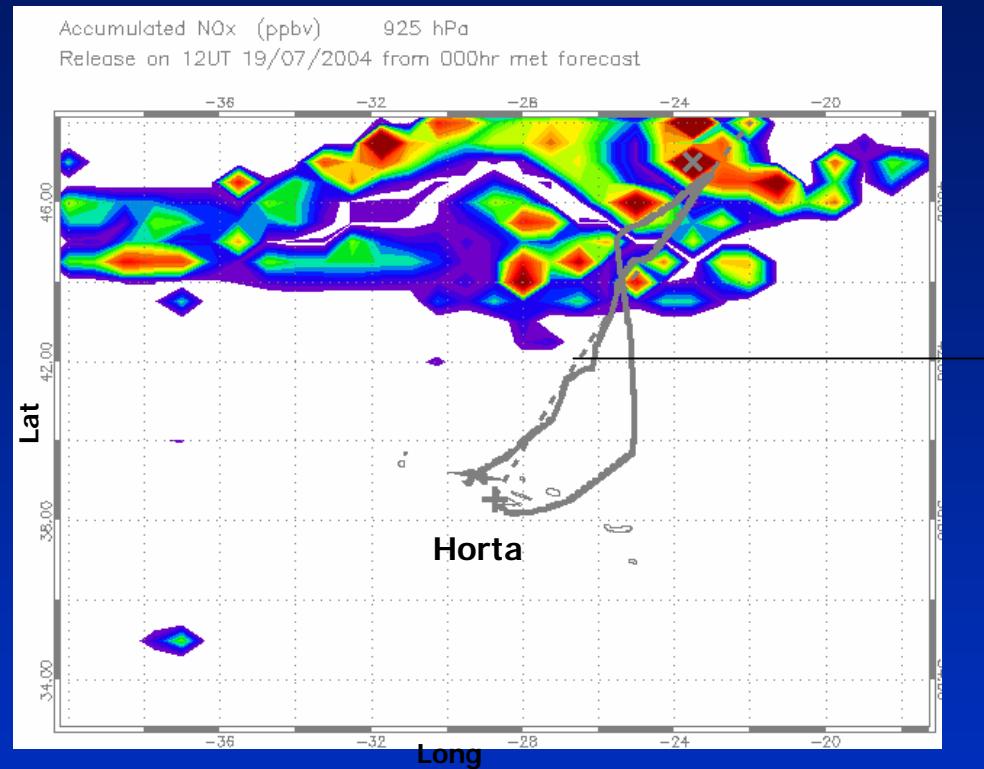
# Forward Trajectories from NOAA P3 flight track



10 day forward trajectories from NOAA P3 flight off NYC on 15/7. Airmass doubles back from Spain to the Azores.

BAE146 flight intercepted this air on 19/7 (saw CO ~ 115-125ppb).

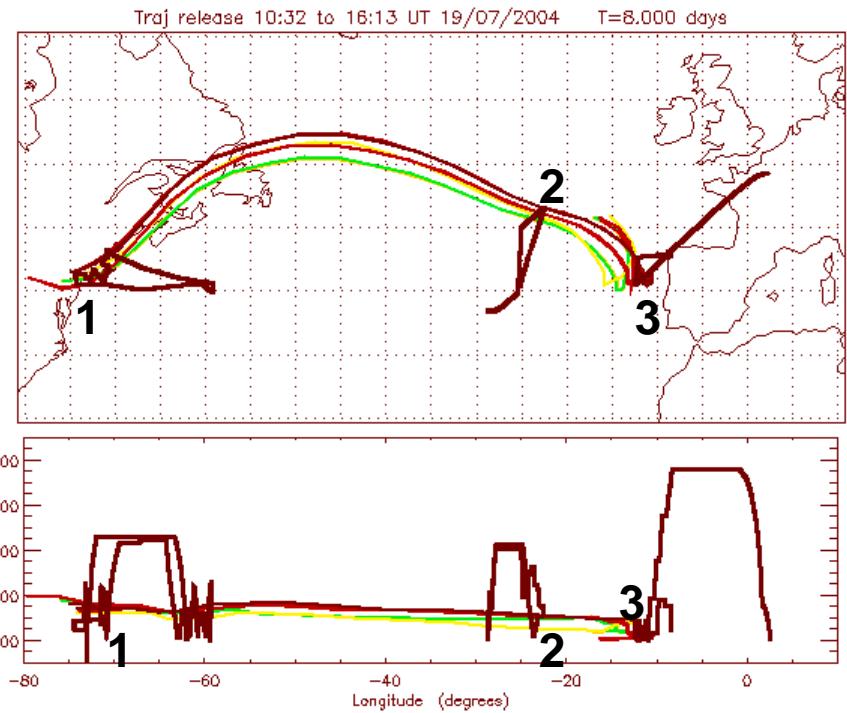
# Analysis: Second interception of polluted airmass from USA



- NO<sub>x</sub> emissions tracer on 925hPa at 12UT 19/7 with BAE146 (air mass relative) flight track overlaid.
- Indicates high emissions into low level outflow from East Coast USA.

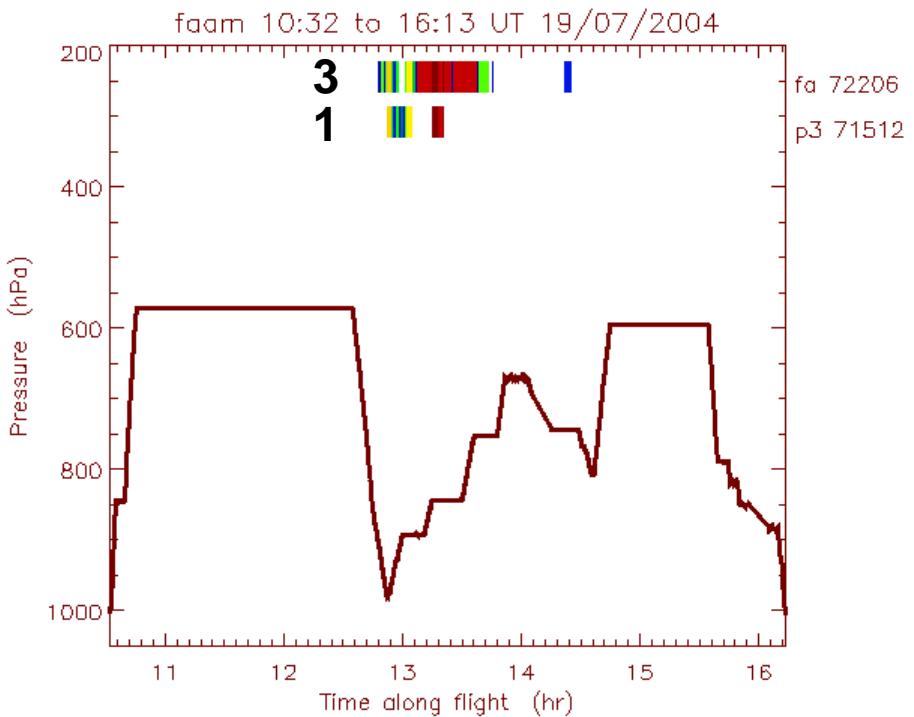
*Back trajectories from target X confirm NYC as being air mass origin*

# Did the aircraft make the interception?



Trajectories from BAe146 flight track back and forwards for 4 days.

Best matches with trajectories from other flight tracks.



Time series along flight track.

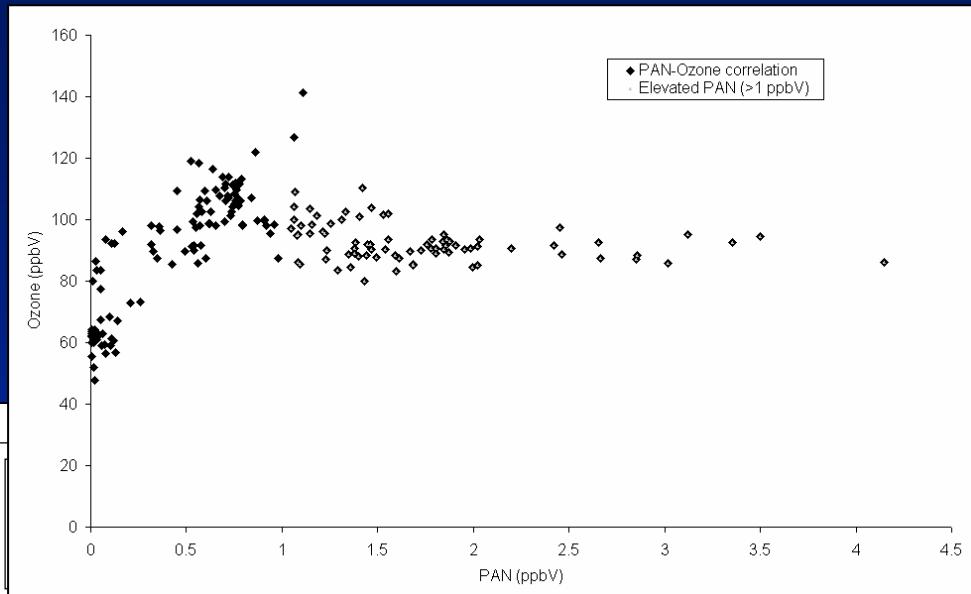
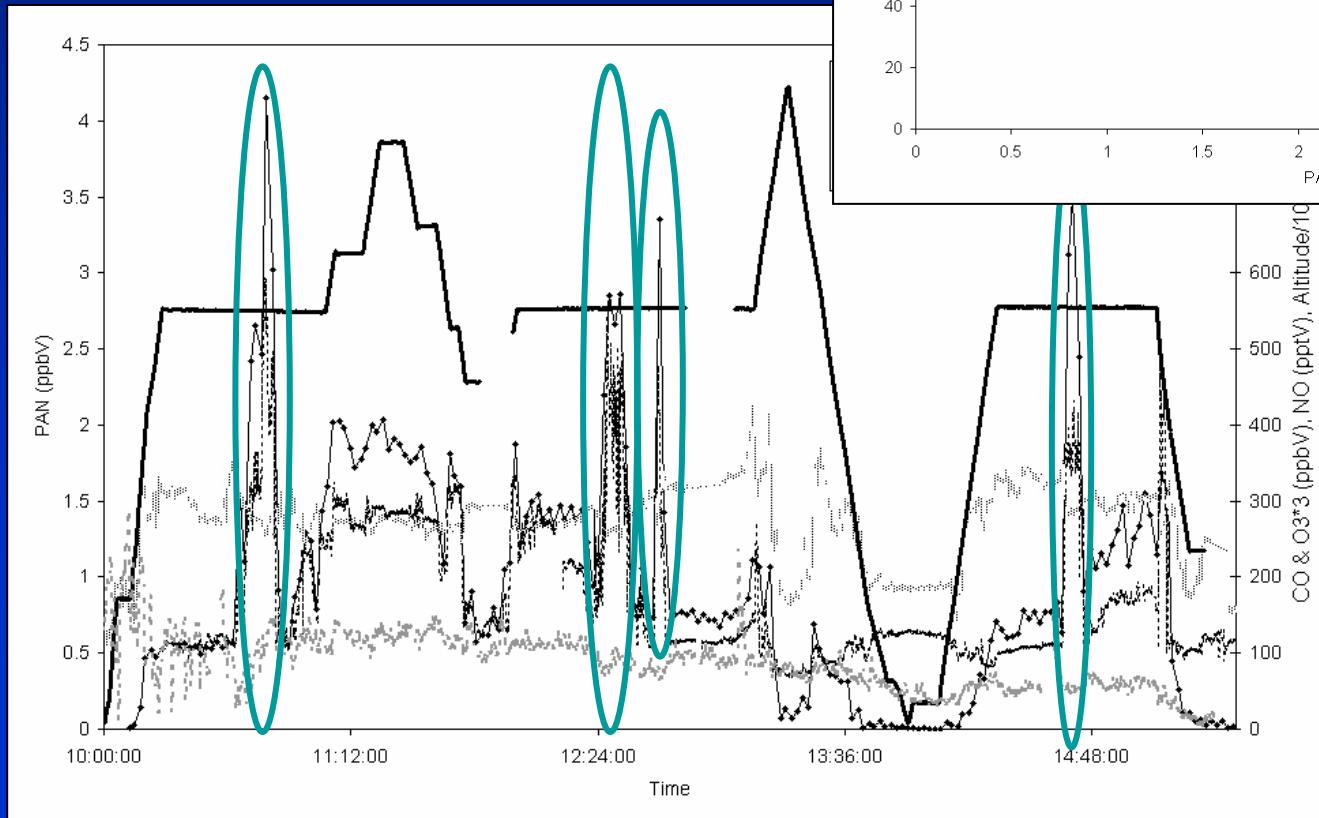
Line shows pressure and colour bars mark air masses intercepted on other flights.

# Was the Quasi-Lagrangian expt successful?

- Trajectories based on analyses show that downstream aircraft flew very close to air masses already sampled upstream.  
But, can it be regarded as the same air mass?
- 1. Do hydrocarbon fingerprints agree (*using ratios to acetylene to allow for mixing and dilution with surrounding air*)?
- 2. Is observed  $\Delta O_3(\tau) >$  instrumental error (*comparison flights*)?  
          > uncertainty in value at origins,  $O_3(t_0 - \tau)$ ?
- 3. What is the sensitivity of  $\Delta O_3$  to initial conditions, reaction pathways, mixing history, ...?

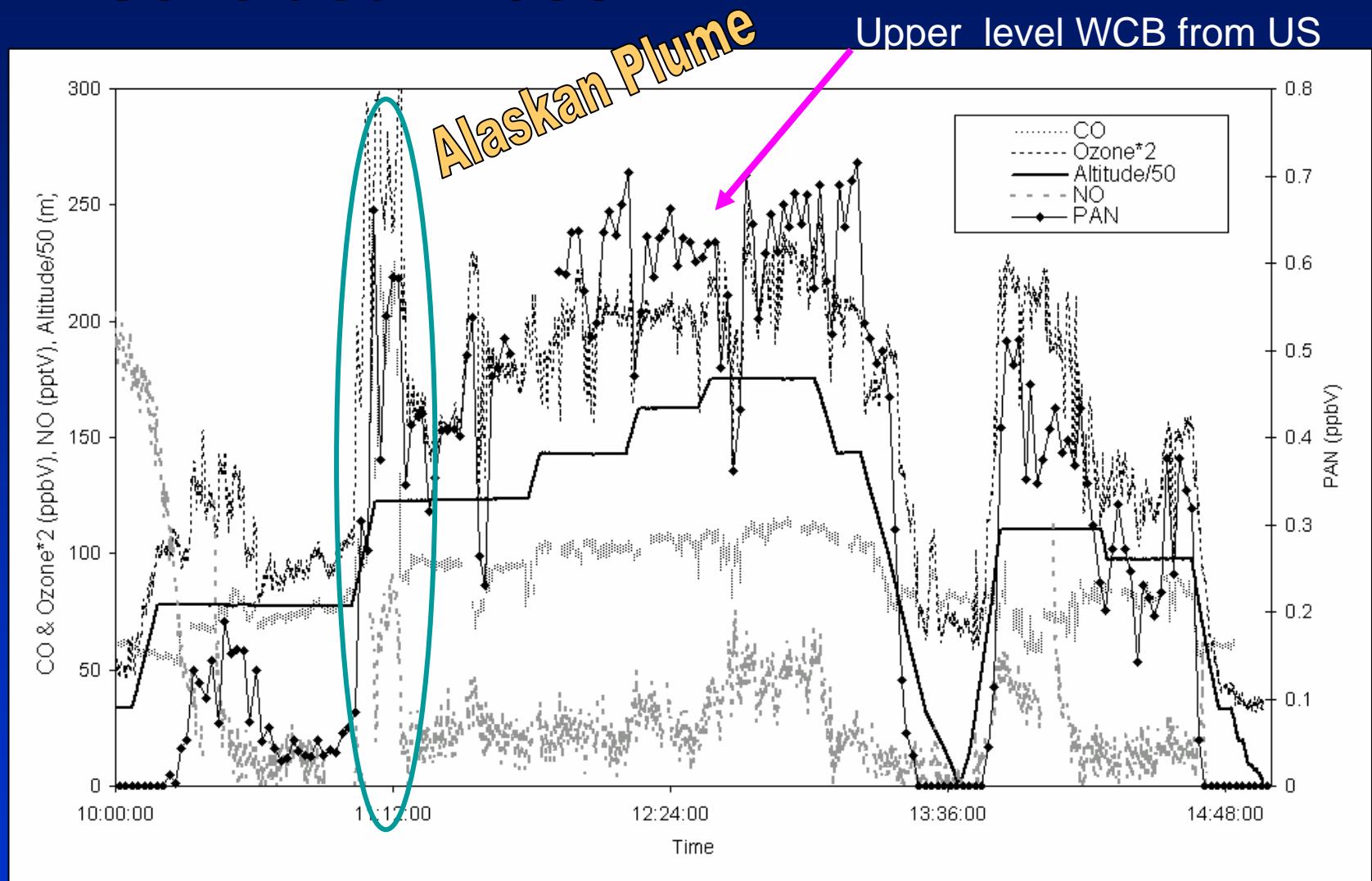
# Alaskan Biomass Plumes: B032

PAN & CO were extremely elevated within the plume and displayed a strong, positive correlation  
Ozone seemed somewhat suppressed within the plume



Alaskan biomass plume was extensively sampled at 5.6 km

# In Contrast...B036



# Plume Comparison

	B032 Plume	B036 Plume
PAN	1.72 ppbV	0.43 ppbV
O <sub>3</sub>	92 ppbV	133 ppbV
CO	261.2 ppbV	132.8 ppbV
(PAN/NOy)*100	39 %	15 %
Temperature	263.4 K	268.0 K

# *Work in progress*

Lagrangian matching using tracers + trajectories. (*Reading / Leeds / York*)

PAN / ozone model studies. (*Leeds / York*)

3D model diagnostics of ozone (*Cambridge*)

Box model / trajectory calculations of o-VOC production (*Leeds*)

HOx determinants (*Leeds*)

Aerosol composition analysis (*Manchester*)

Lagrangian model to investigate sensitivity of trans-Atlantic  $\Delta O_3$  to:

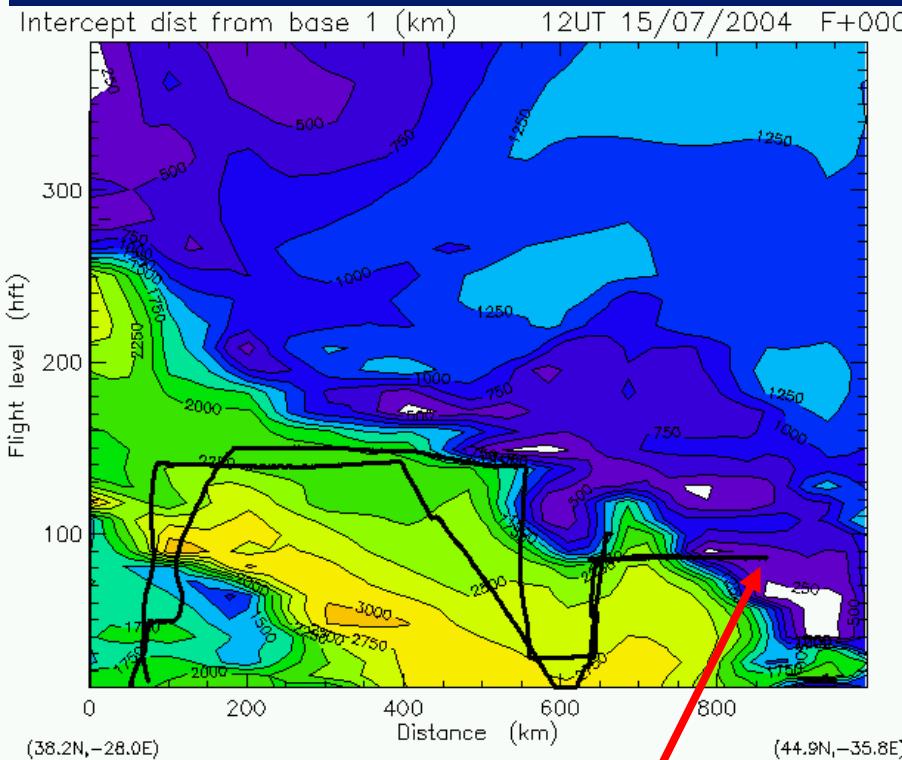
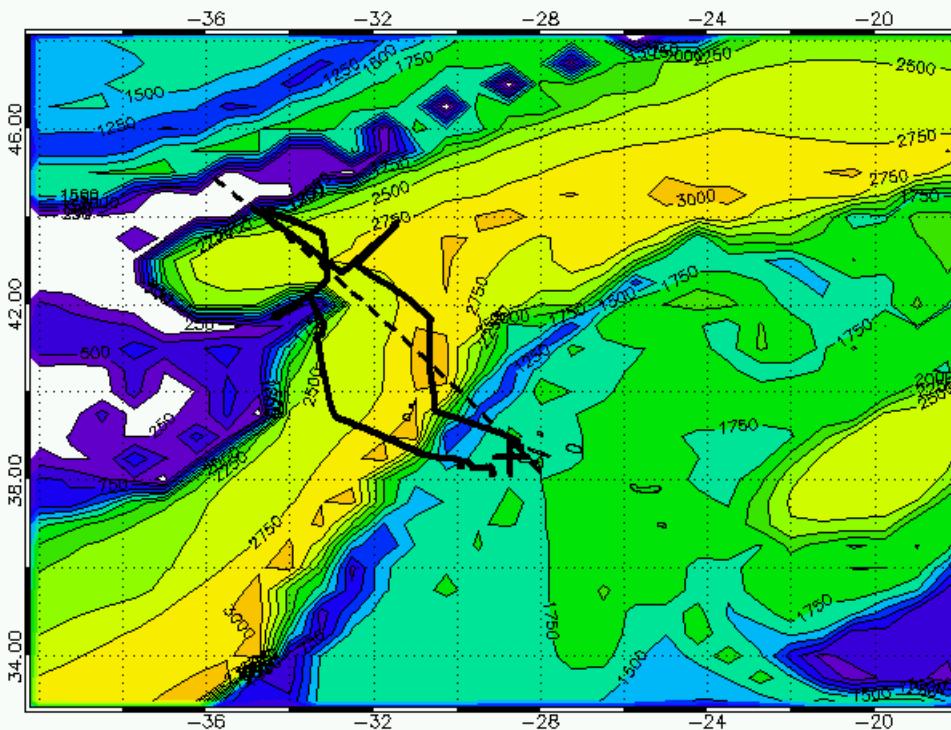
- *Mixing rate*
- *Cloud cover*
- *ECMWF water vapour*
- *NMHC complexity*
- *Initial conditions*
- Constraint of mixing term through hydrocarbons.
- Full Monte-Carlo analysis using Lagrangian model.
- Can reduced chemical mechanisms describe the observed change in composition or is complexity unavoidable?



# **UK ITOP flight by flight summary**

# B029: 15/07/04 Low level pollution and fires

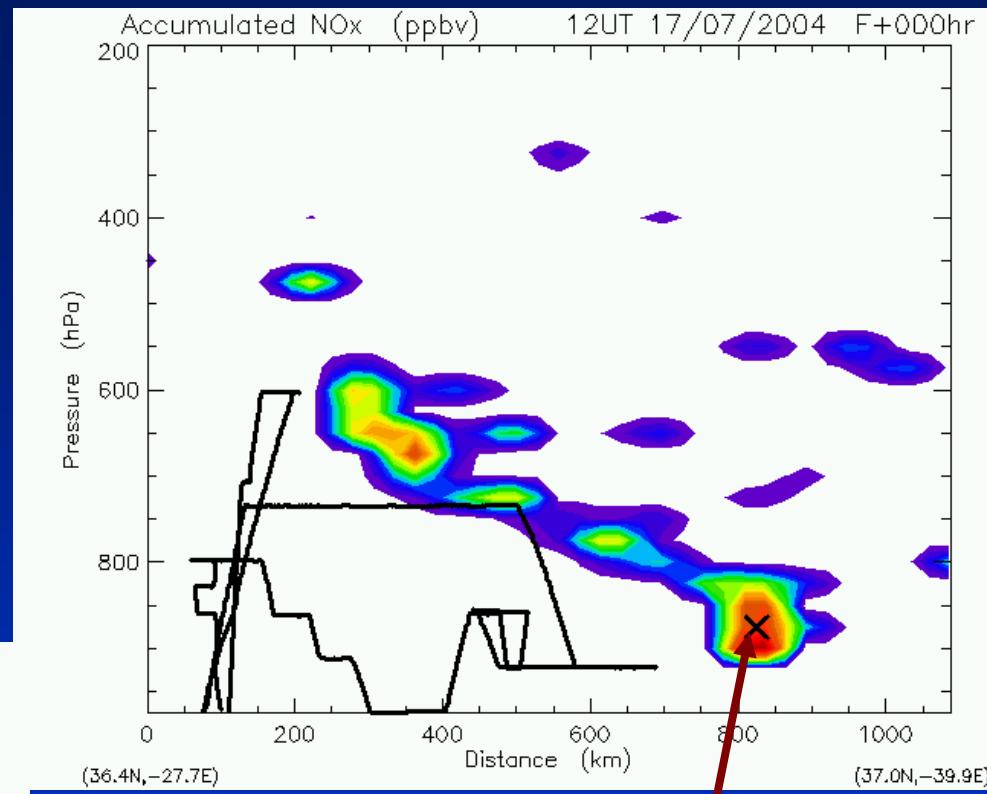
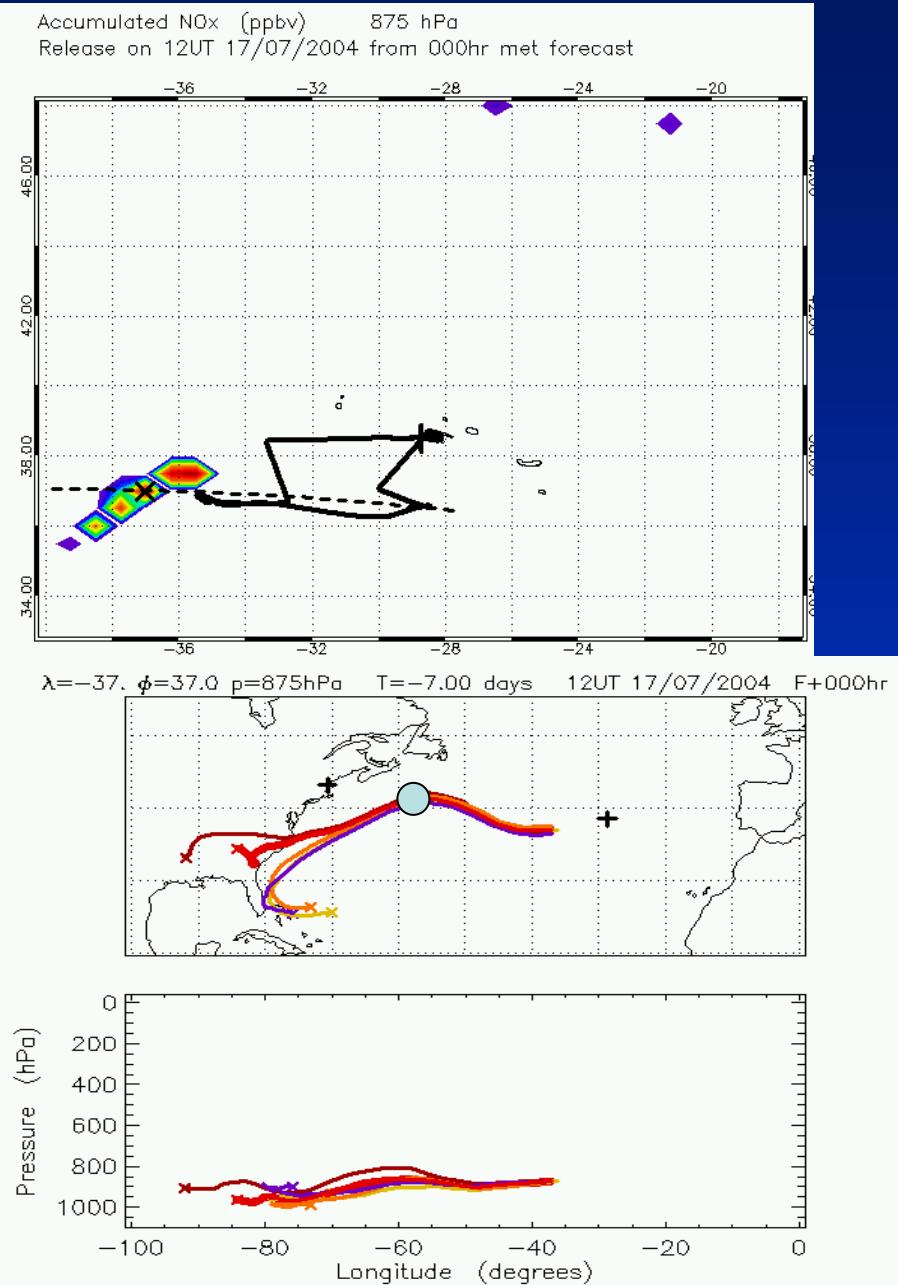
Intercept dist from base 1 (km) 850 hPa  
Release on 12UT 15/07/2004 from 000hr met forecast



# Horta

## Air passing close to Pease

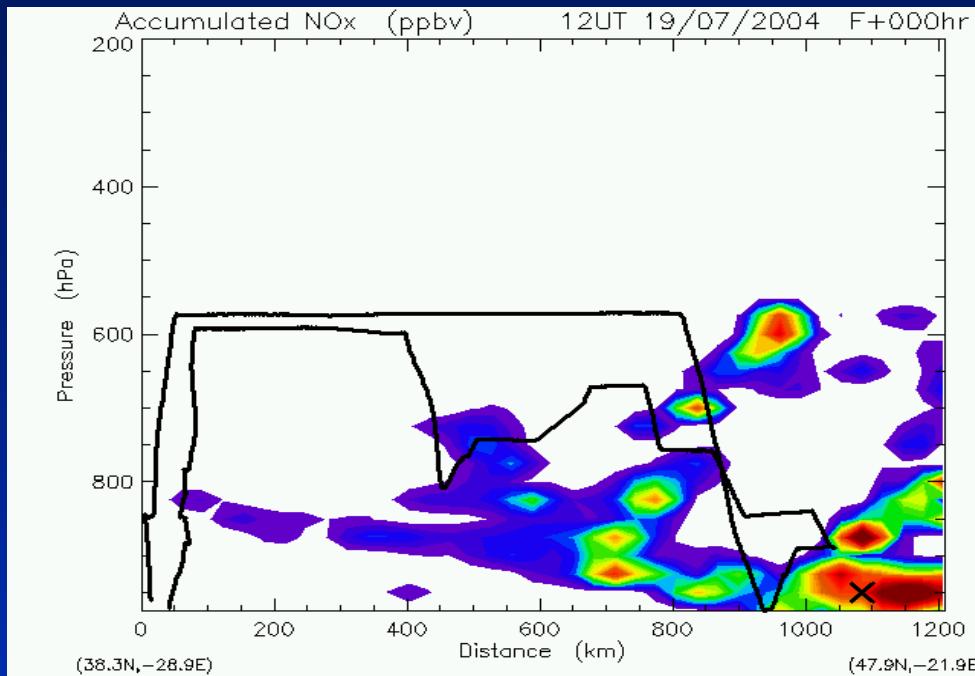
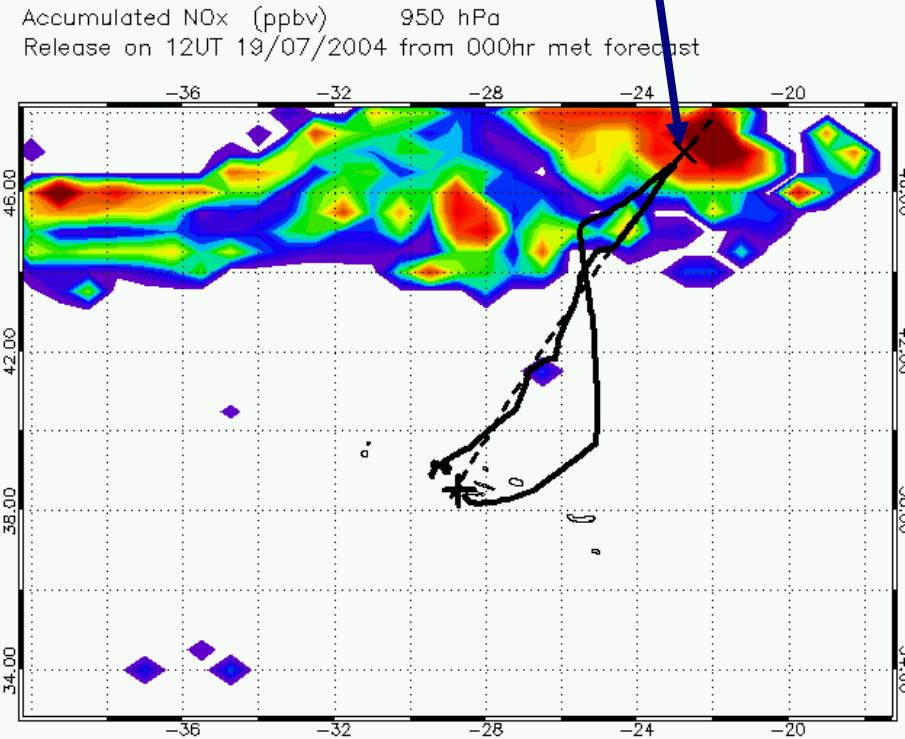
# B030: 17/07/04 First Lagrangian opportunity



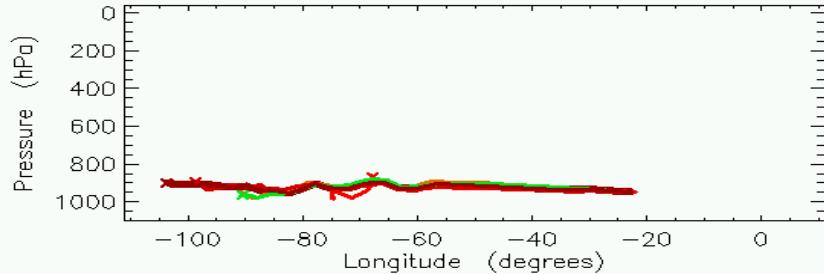
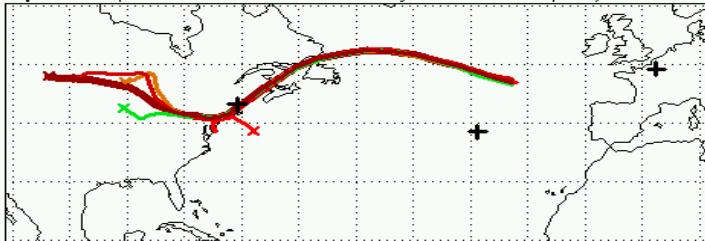
Target sampled by P3 on 15 July  
Unfortunately only skimmed edge because out of time.

# B031: 19/07/04 “New York plume”

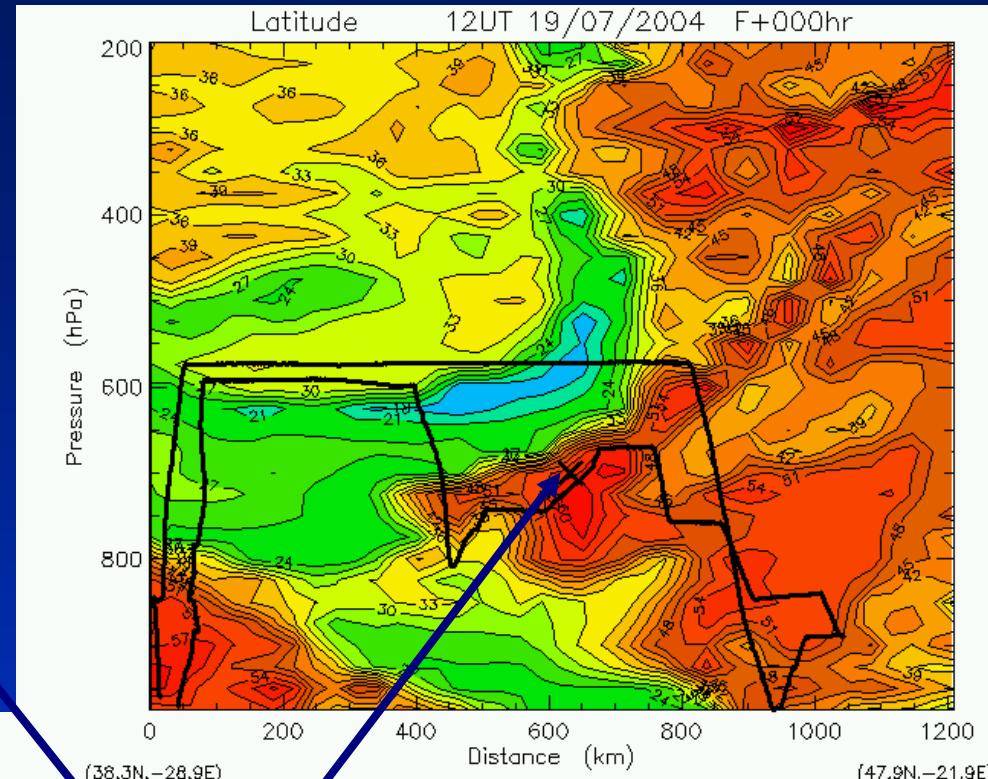
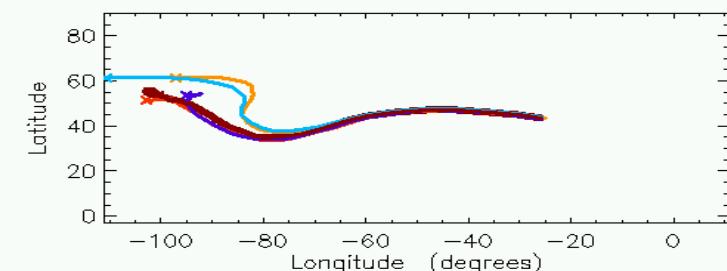
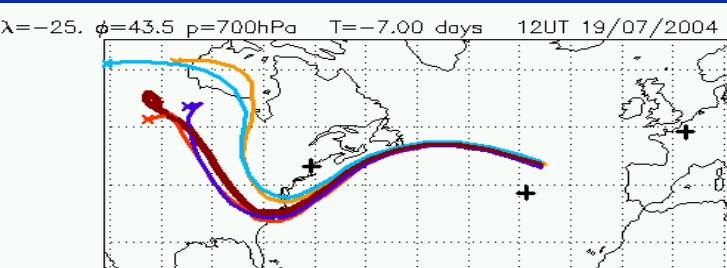
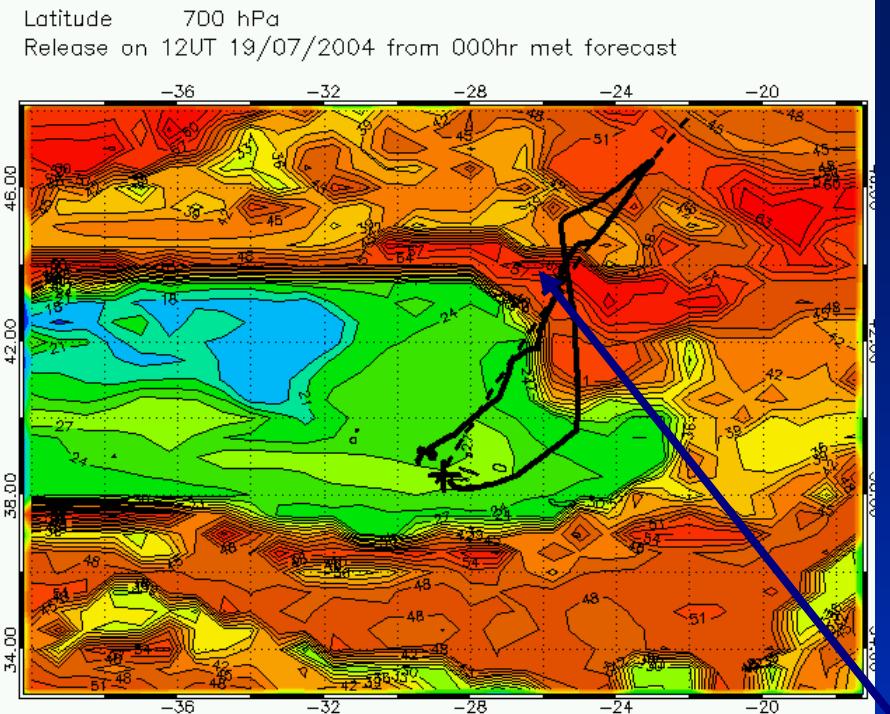
Target sampled by P3 on 15 July (on limit of range)



$\lambda = -22$ ,  $\phi = 47.0$ ,  $p = 950 \text{ hPa}$ ,  $T = -7.00 \text{ days}$ , 12UT 19/07/2004, F+000hr



# B031 continued: thin brown fire layer

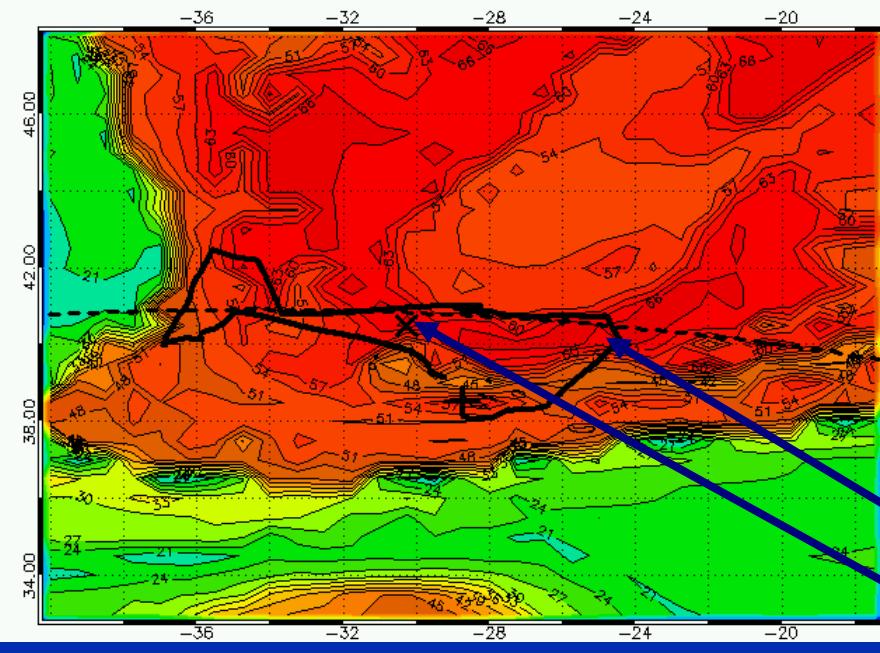


Narrow filament and thin layer - visibly brown.  
Adjusted altitude to stay within layer.

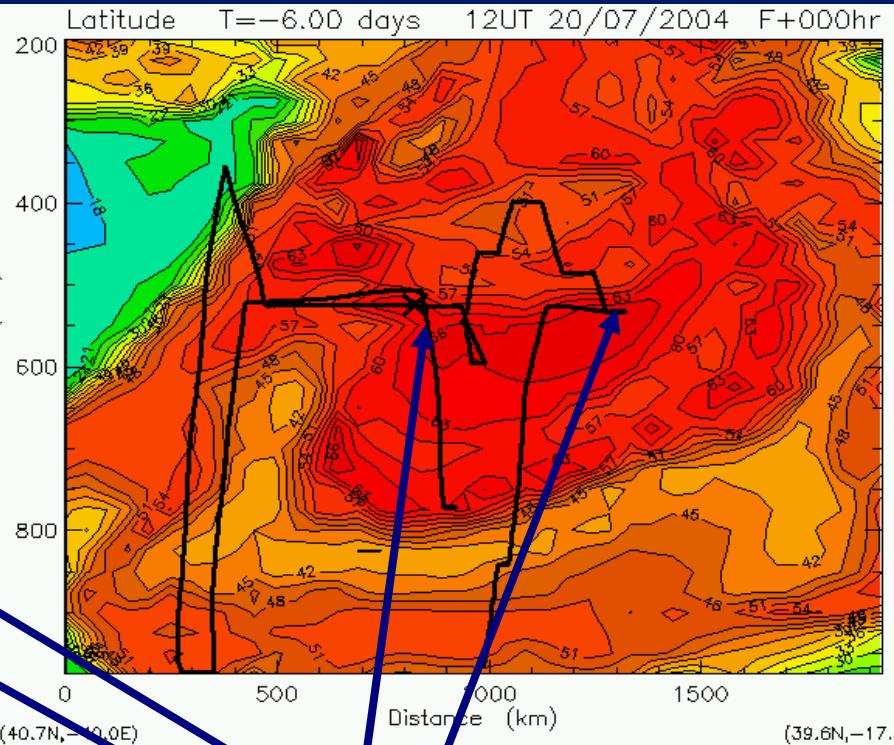
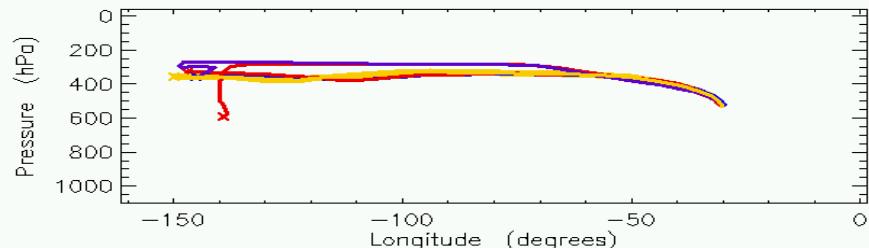
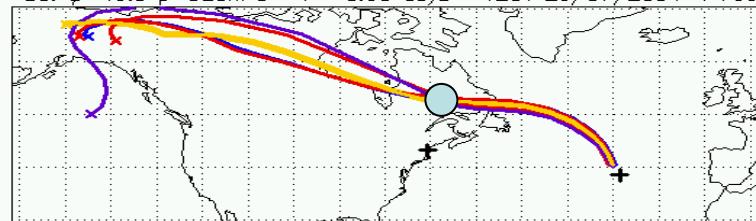
Originated from UT above Alaskan forest fires  
(but diluted by mixing while thinning).

# B032: 20/07/04 Alaskan forest fires

Latitude T=-6.00 days 525 hPa  
 Release on 12UT 20/07/2004 from 000hr met forecast



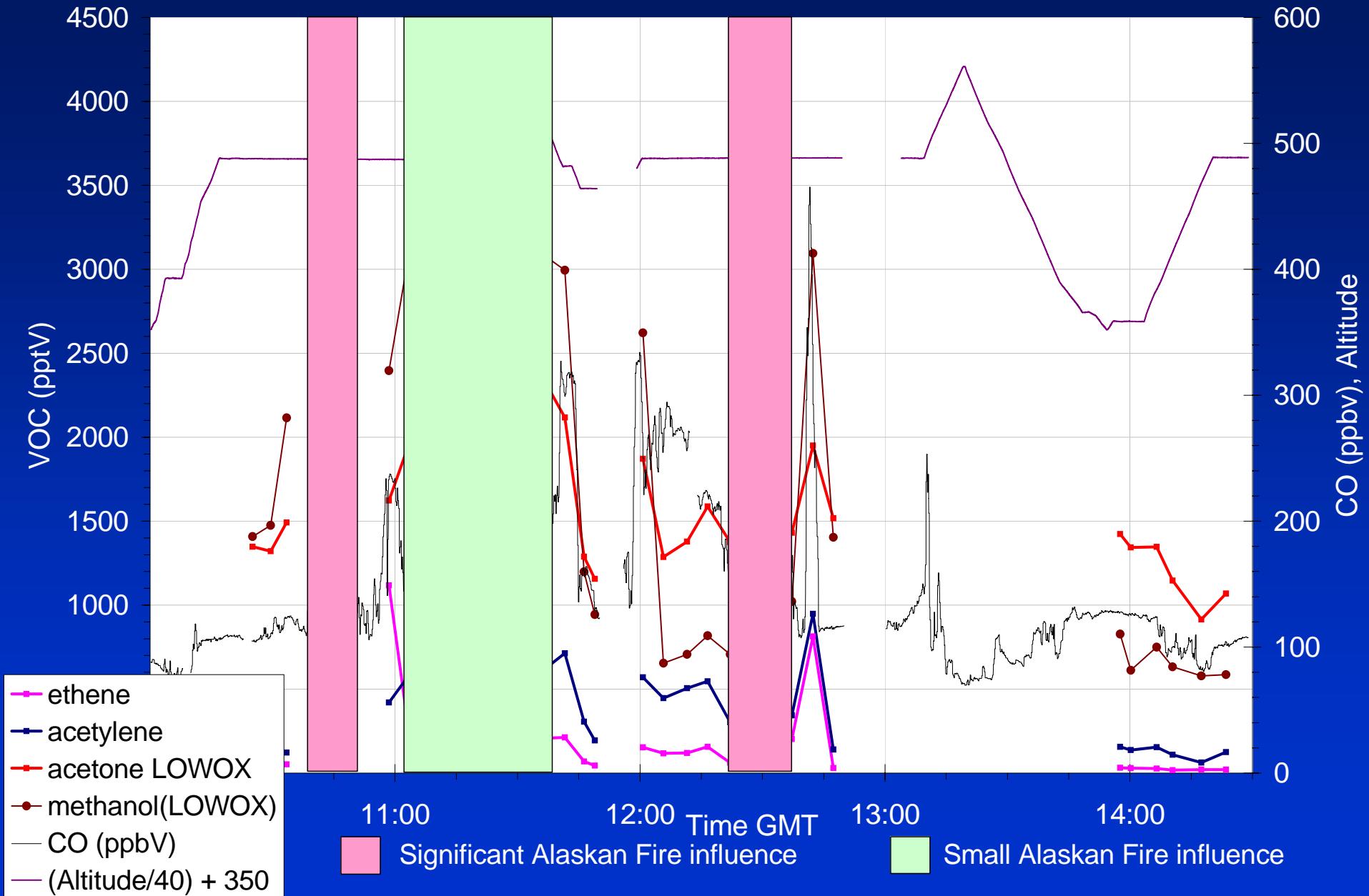
$\lambda = -30^\circ$ ,  $\phi = 40.5^\circ$ ,  $p = 525\text{ hPa}$ ,  $T = -6.00$  days, 12UT 20/07/2004, F+000hr



Alaskan fire plume with CO up to 600 ppb.  
 Similar plume was intercepted by P3 on 15 July (but not same air mass).

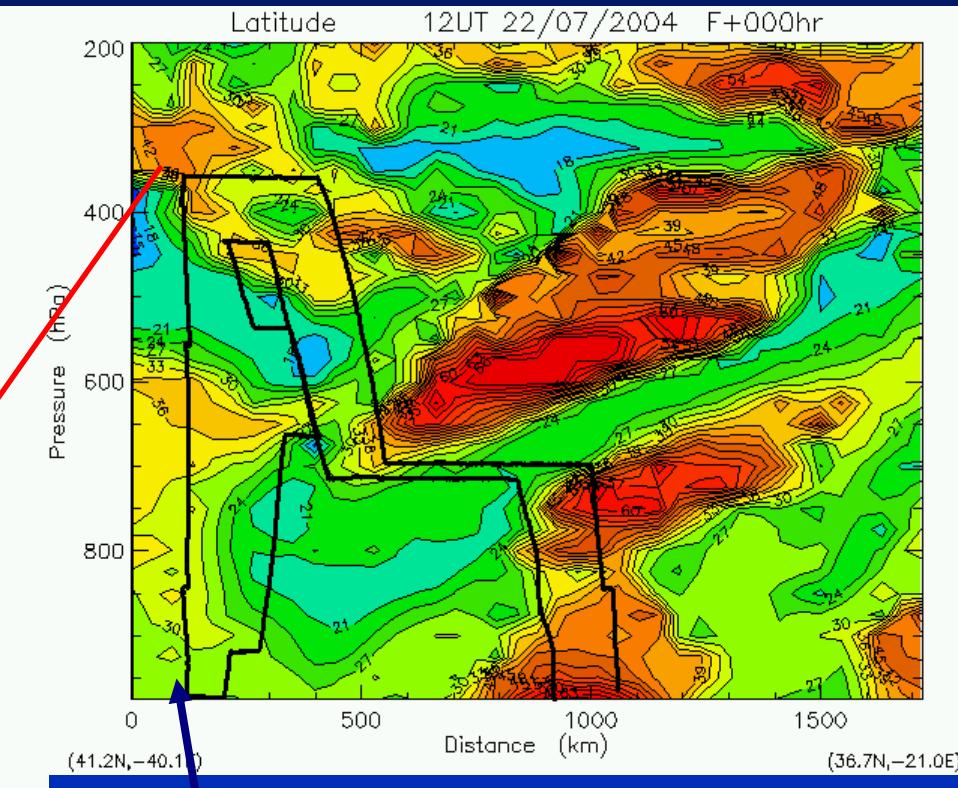
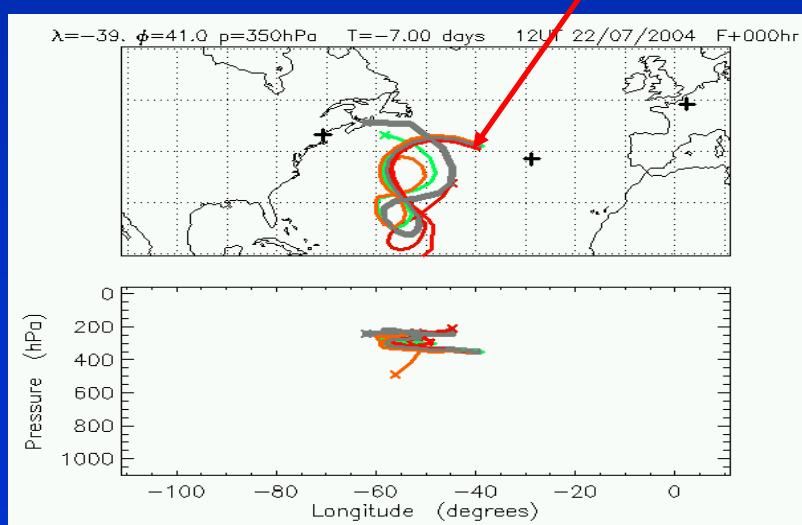
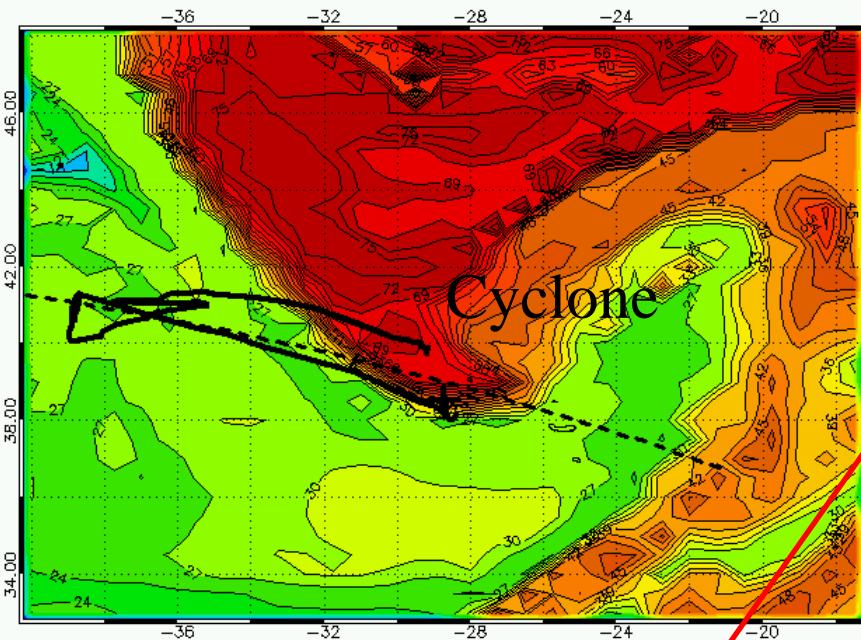
Was targeted on 23/7 by Falcon.

# B032 NHCMS



# B033: 22/07/04 ENVISAT underpass

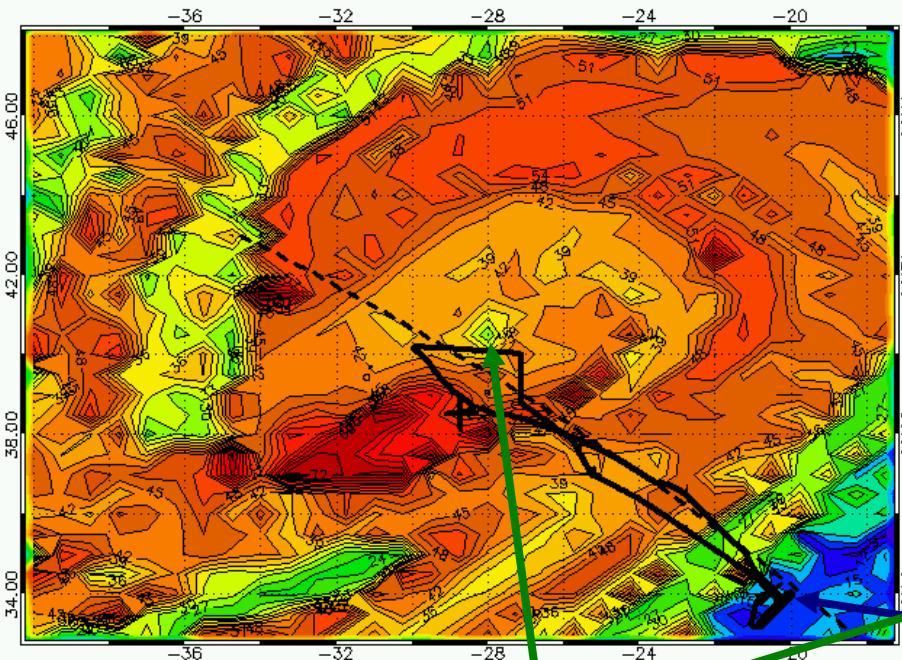
Latitude 975 hPa  
Release on 12UT 22/07/2004 from 000hr met forecast



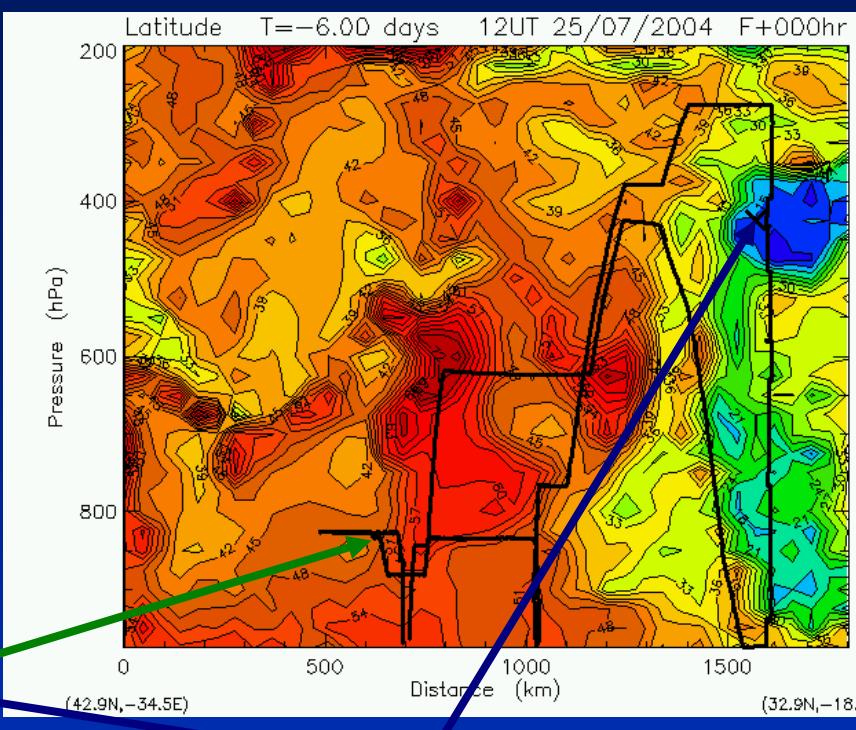
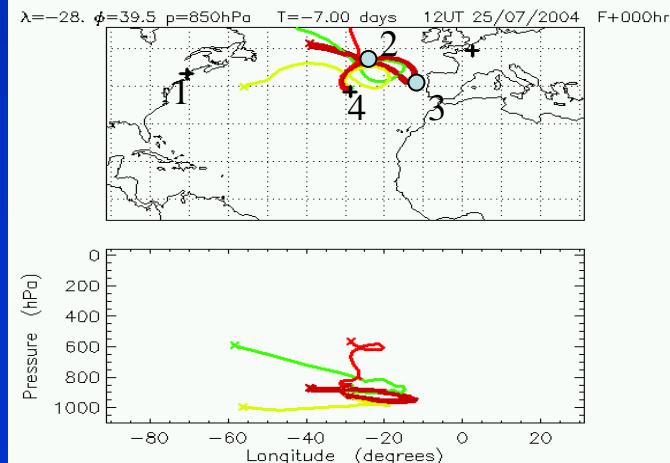
**Deep profile under satellite through air re-circulating over mid-Atlantic for more than a week.**

# B034: 25/07/04 African air

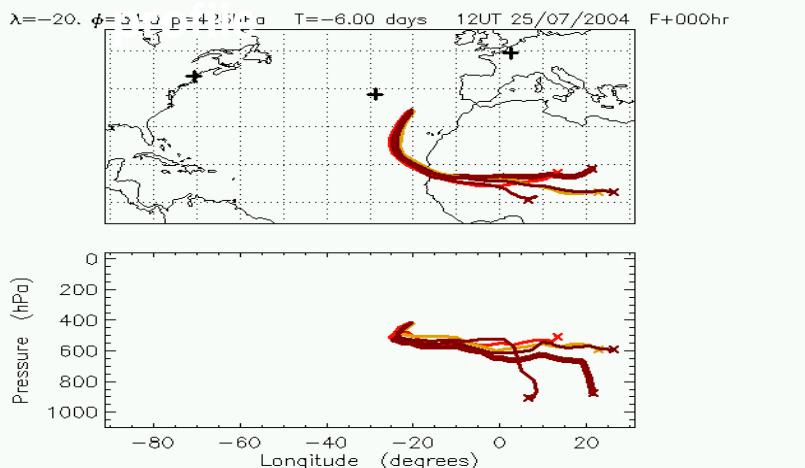
Latitude T=-6.00 days 425 hPa  
 Release on 12UT 25/07/2004 from 000hr met forecast



New York plume 4<sup>th</sup> interception

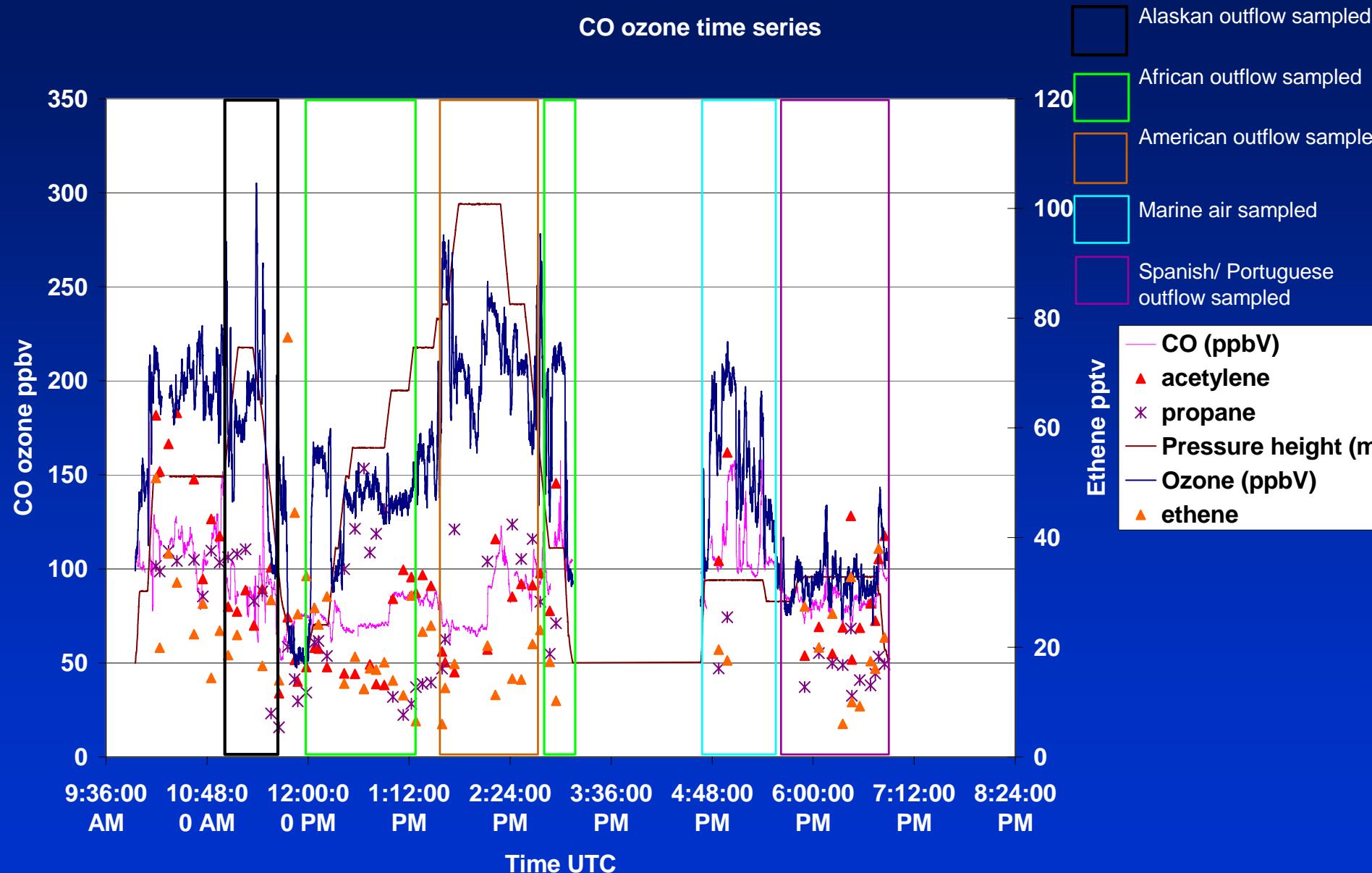


African air throughout



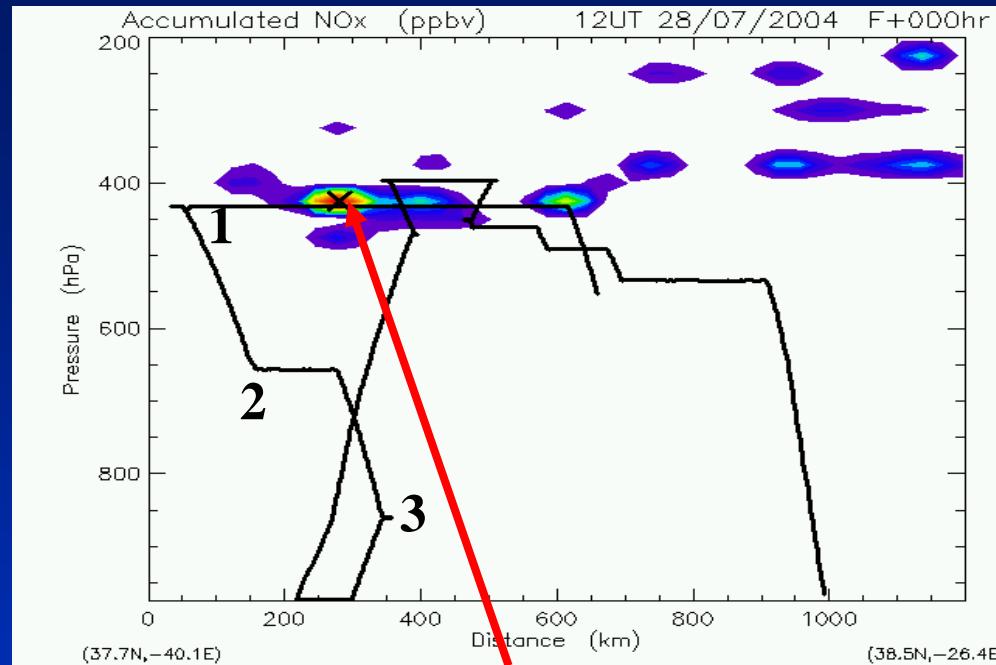
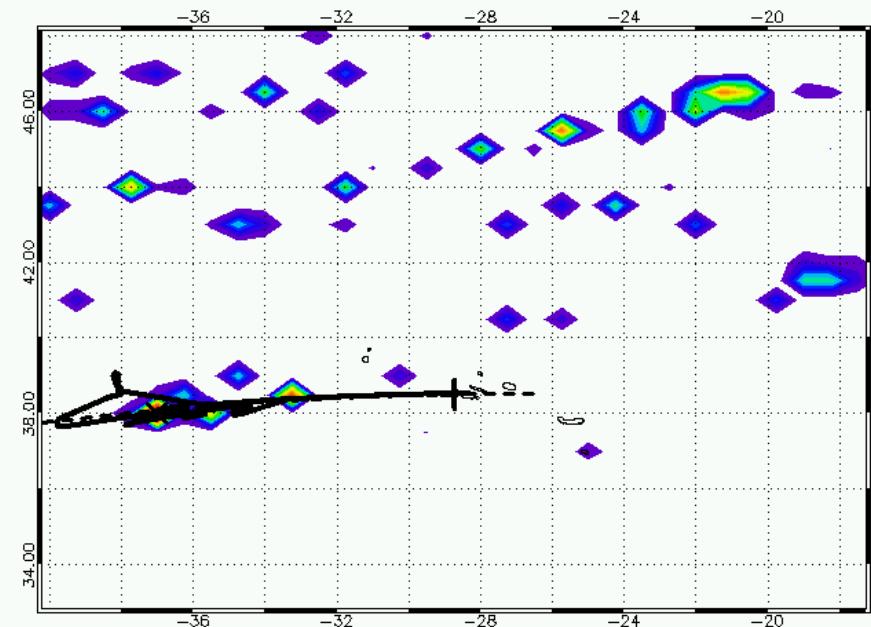
# B034: 25/07/04 African air

## CO ozone time series

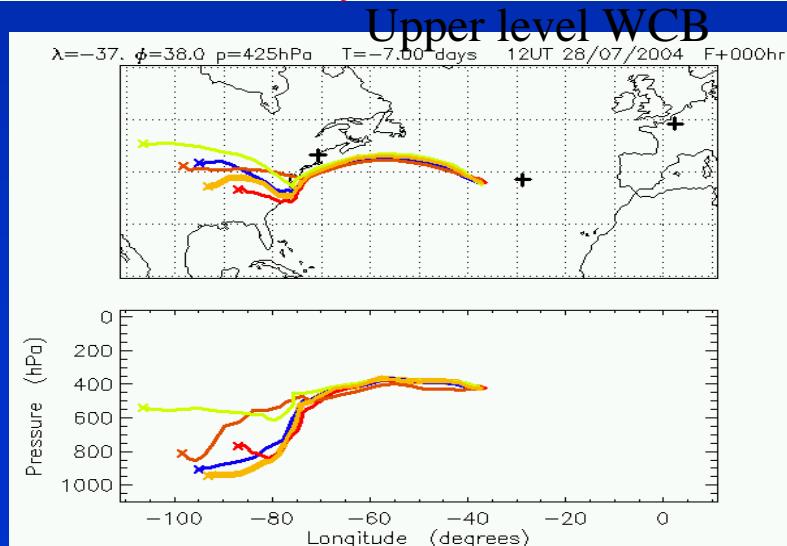


# B035: 28/07/04 Comparison with DC8

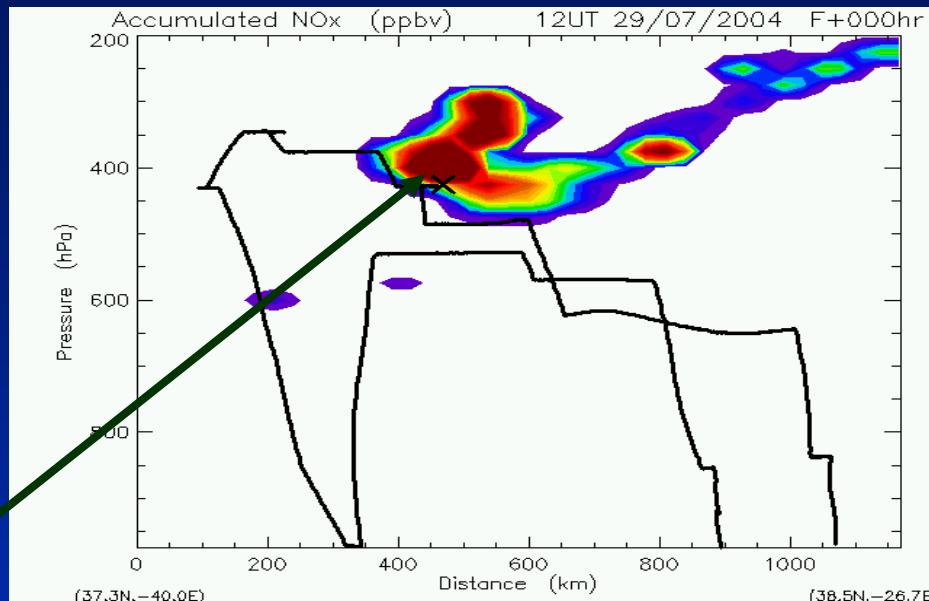
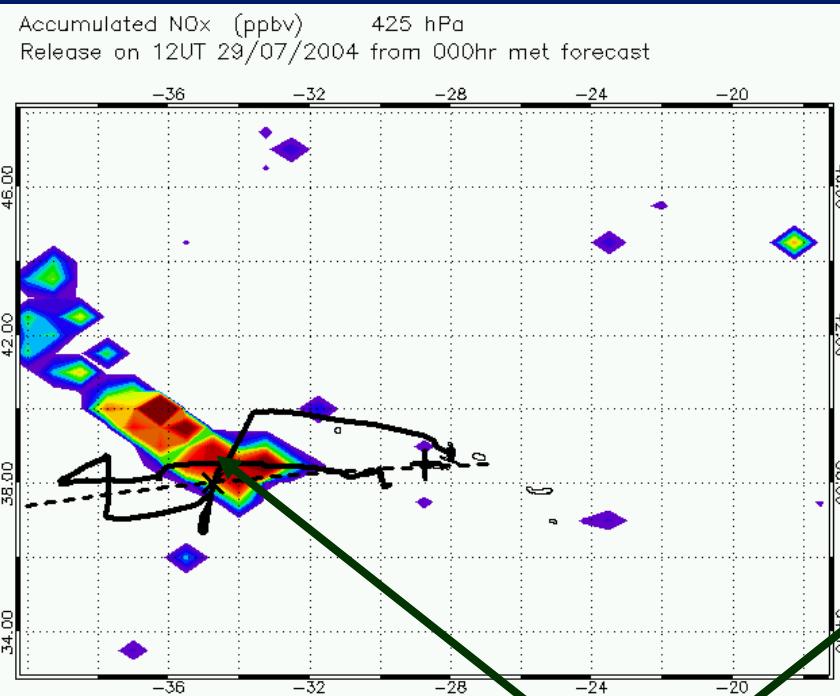
Accumulated NO<sub>x</sub> (ppbv) 425 hPa  
Release on 12UT 28/07/2004 from 000hr met forecast



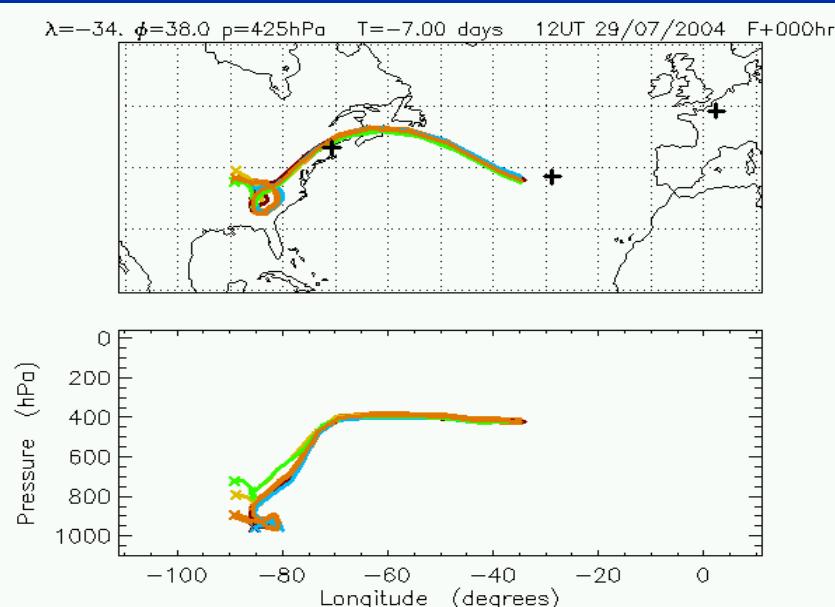
**Comparison below air exported by WCB. Relatively clean but sampled again by Falcon on 30/7 and 31/7.**



# B036: 29/07/04 Upper level export in WCB

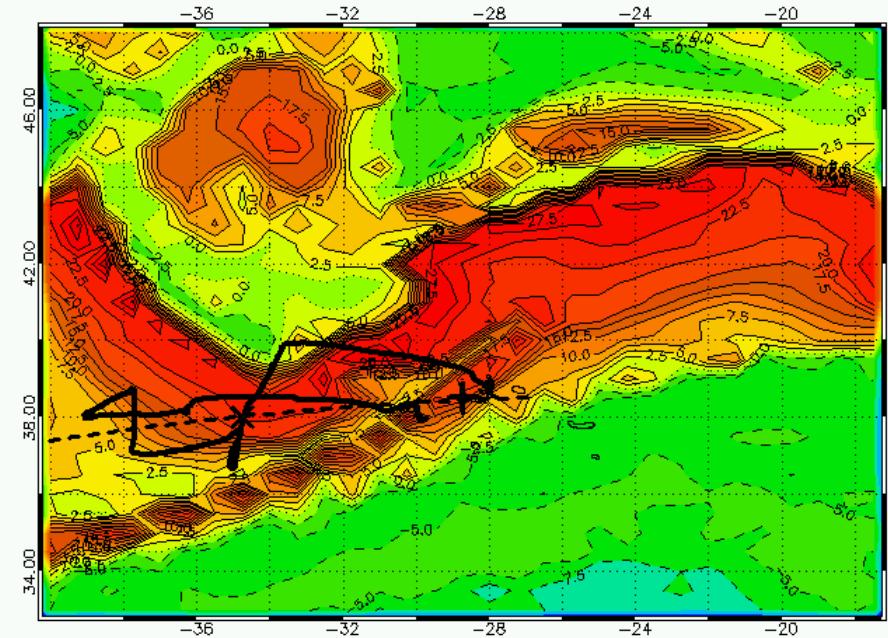


**Polluted air on North side of WCB but crossed under maximum because could not achieve altitude early enough.**



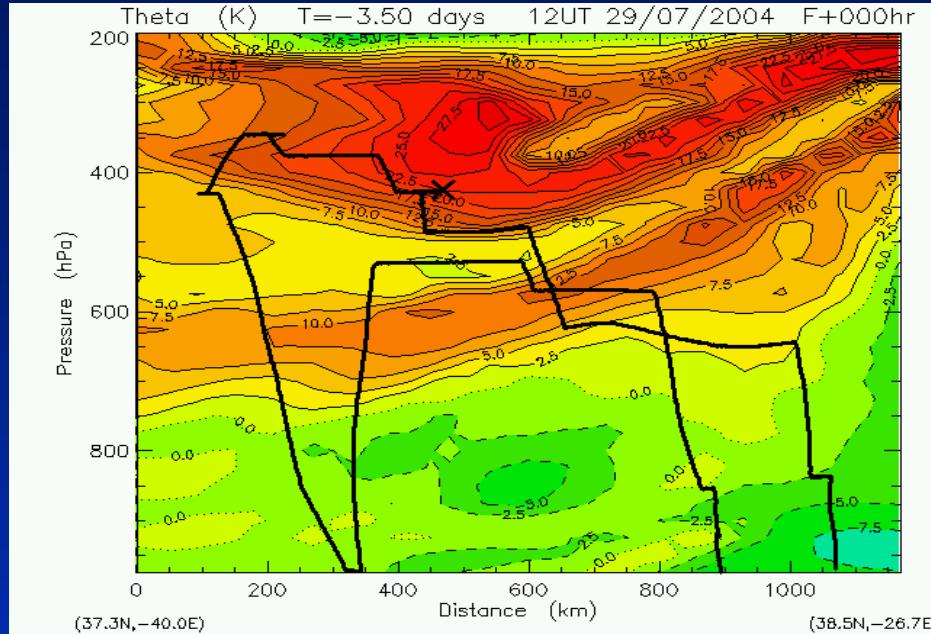
# B036 continued: WCB features

Theta (K) T=-3.50 days 425 hPa  
Release on 12UT 29/07/2004 from 000hr met forecast

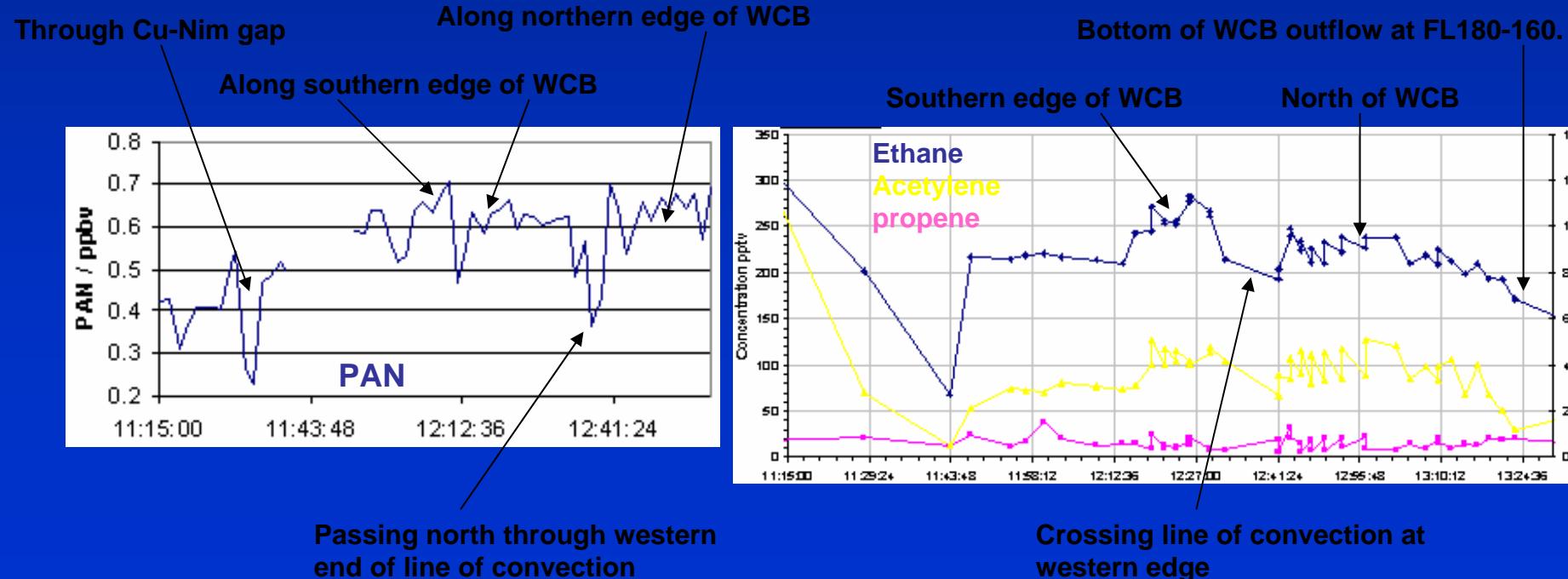
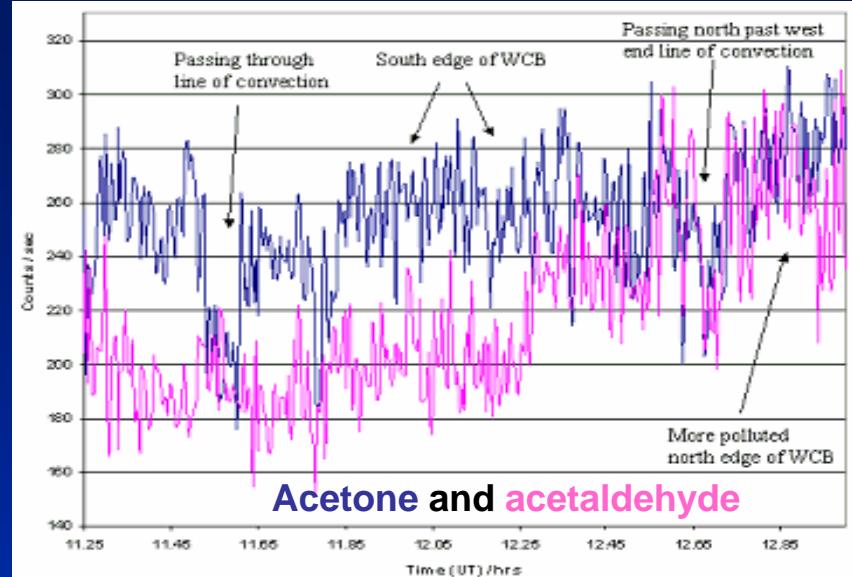
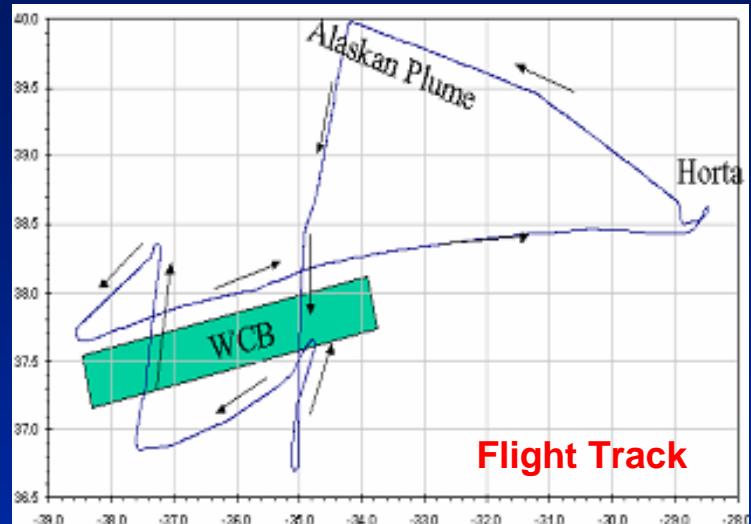


**Observed line of convection  
(dashed line) was parallel to  
feature lying to south at upper  
levels (figure above)**

**Narrow line of deep convection  
– BA146 flew along south side  
and then a deep profile on north  
side**

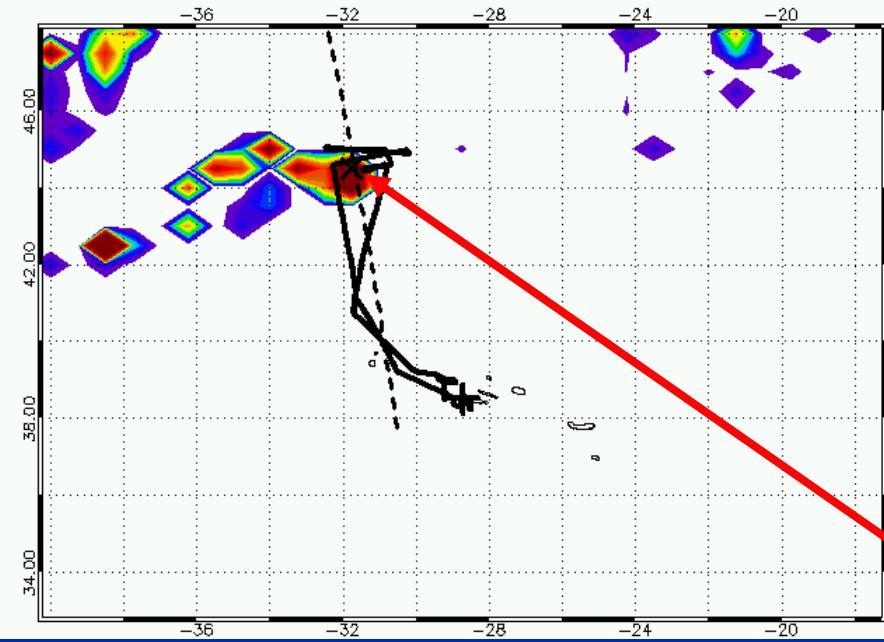


# Flight B036

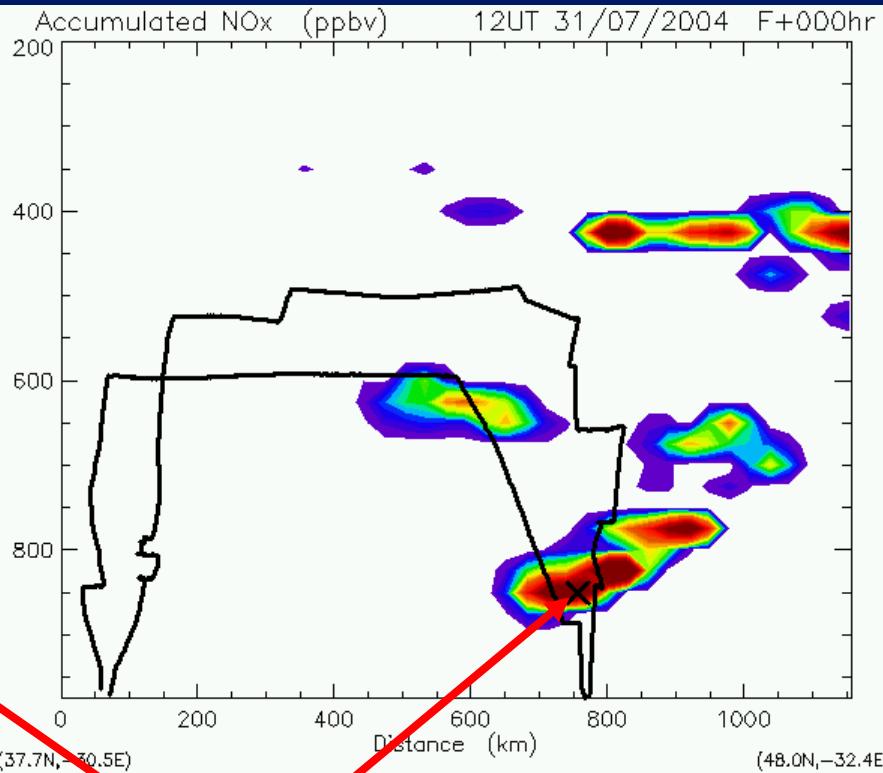
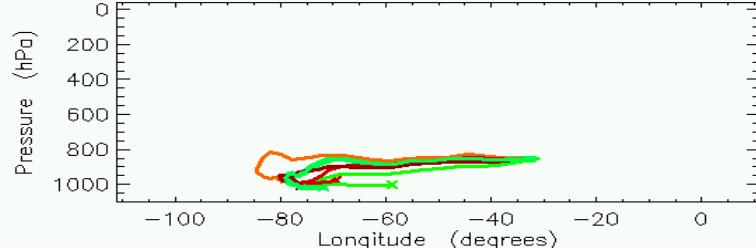
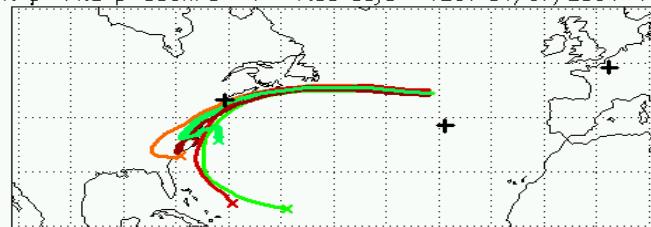


# B037: 31/07/04 Low level WCB (1)

Accumulated NOx (ppbv) 850 hPa  
Release on 12UT 31/07/2004 from 000hr met forecast

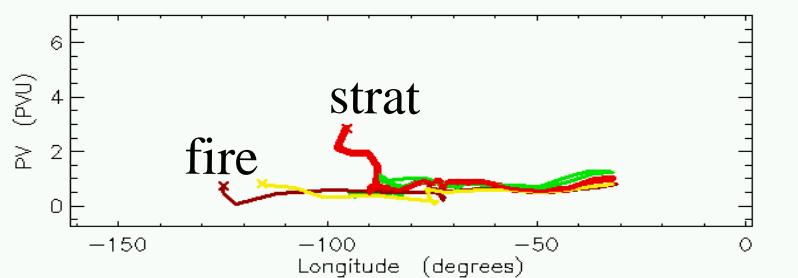
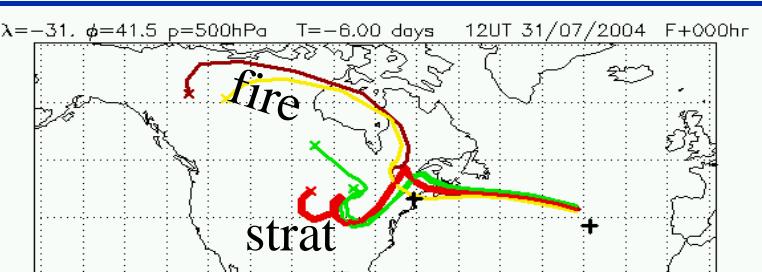
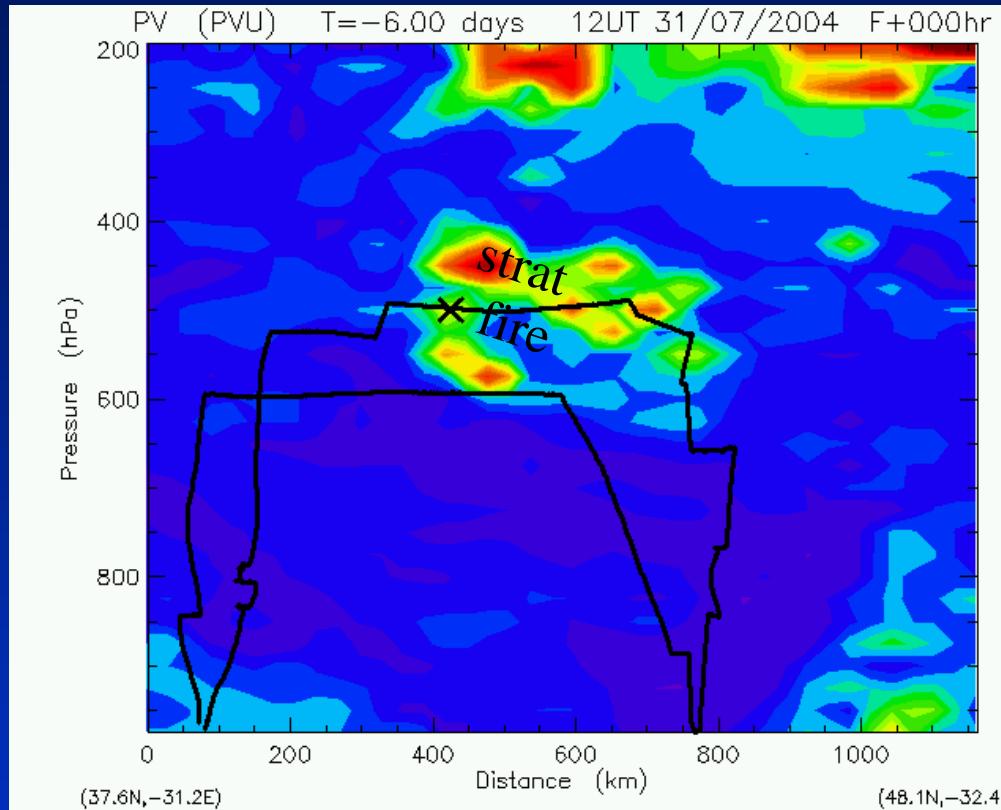
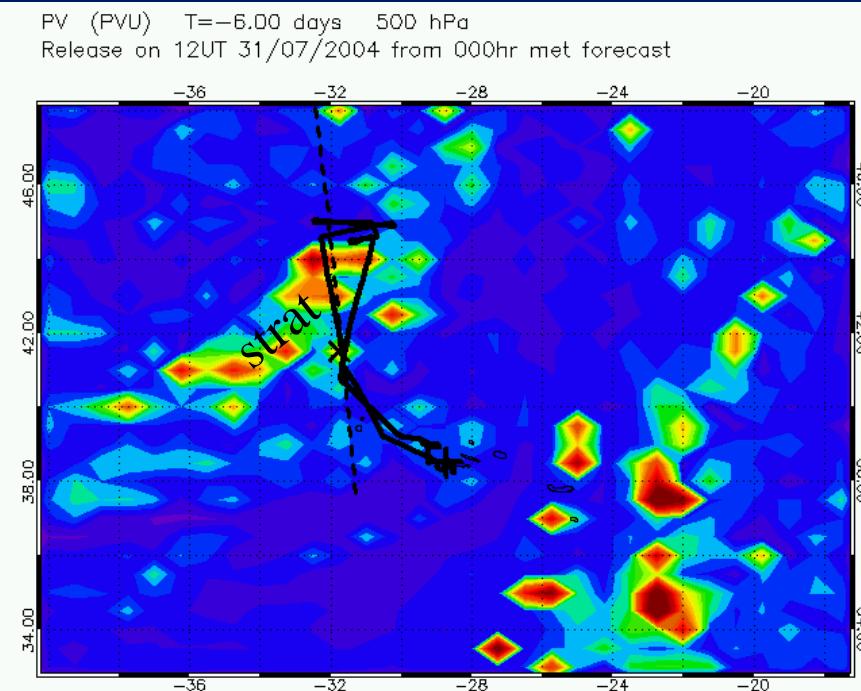


$\lambda = -31$ ,  $\phi = 44.5$ ,  $p = 850 \text{ hPa}$ ,  $T = -7.00 \text{ days}$ , 12UT 31/07/2004, F+000hr



Target already sampled by P3 on 27 and 28 July

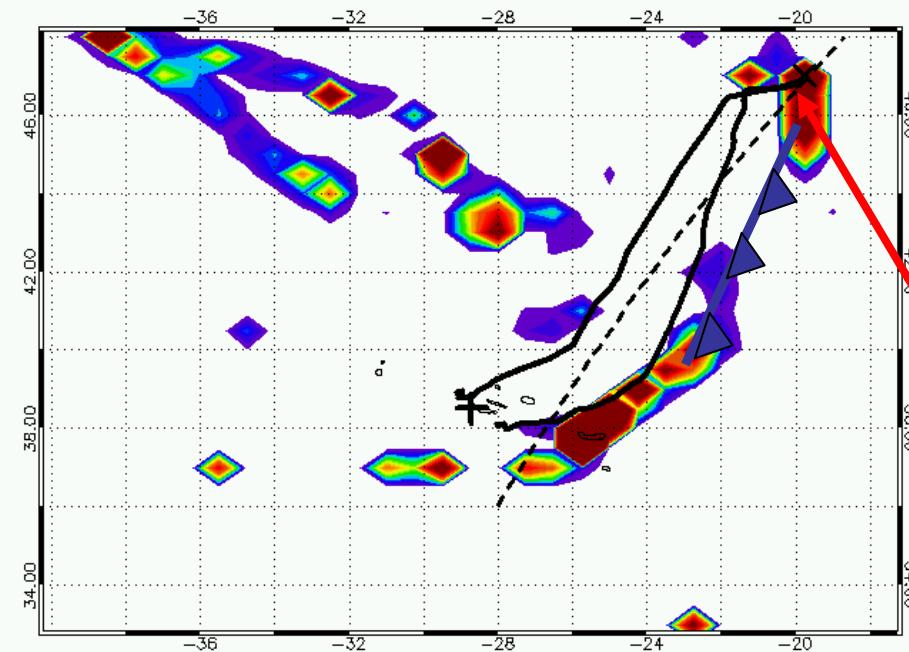
# B037 continued: forest fire mixed with stratospheric air



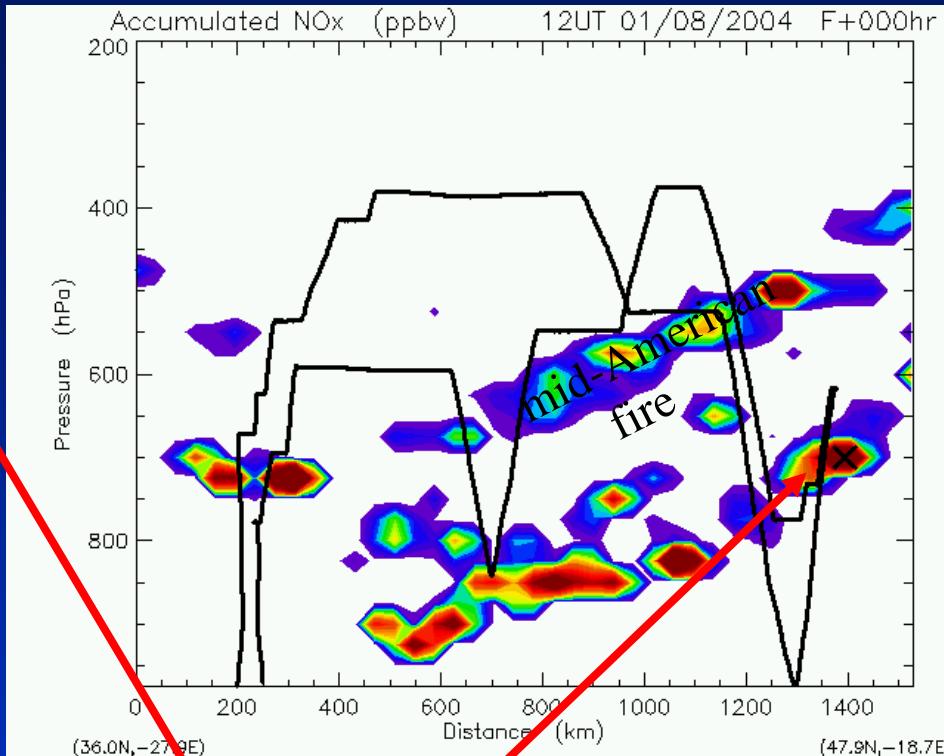
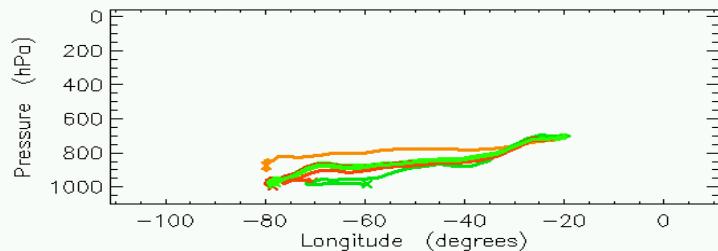
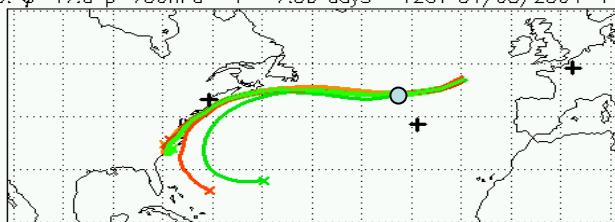
**Strong evidence for forest fire CO mixed with stratospheric ozone**

# B038: 01/08/04 Low level WCB (2)

Accumulated NOx (ppbv) 700 hPa  
Release on 12UT 01/08/2004 from 000hr met forecast

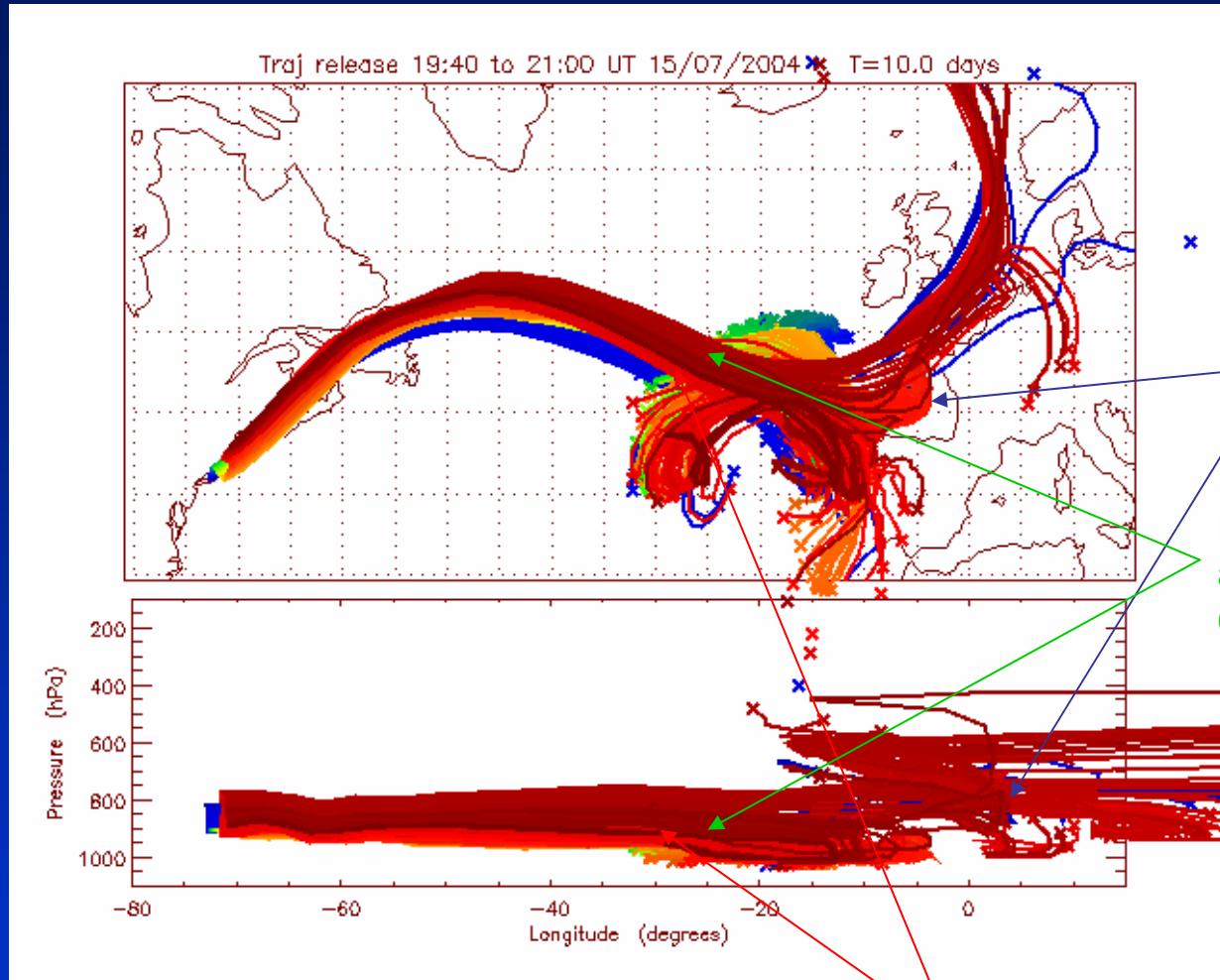


$\lambda = -19$ ,  $\phi = 47.0$ ,  $p = 700\text{hPa}$ ,  $T = -7.00$  days, 12UT 01/08/2004, F+000hr



Rapidly receding target  
already sampled by P3 on 27  
and 28 July and BAe146 on 31  
July.

# Possible multiple interception of New York polluted airmass: 1



10 day forward trajectories from NOAA P3 flight off NYC on 15/7. Airmass doubles back from Spain to the Azores.

BAE146 flight intercepted this air on 19/7 (saw CO ~ 115-125 ppb).

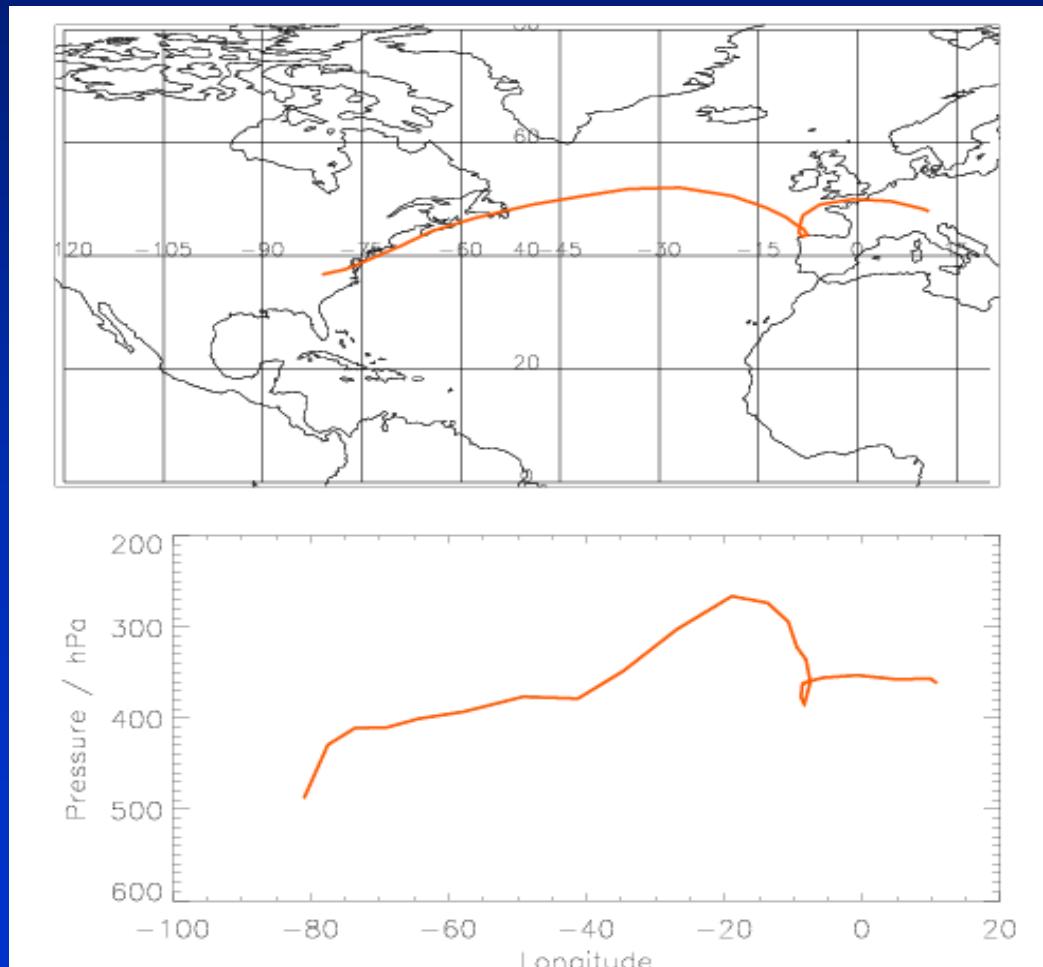
Intercepted by DLR Falcon off NW corner of Spain on 22/7 (saw CO ~ 90-100 ppb).

BAE146 flight intercepted this air again on 25/7 (saw increased CO ~ 115-125 ppb).

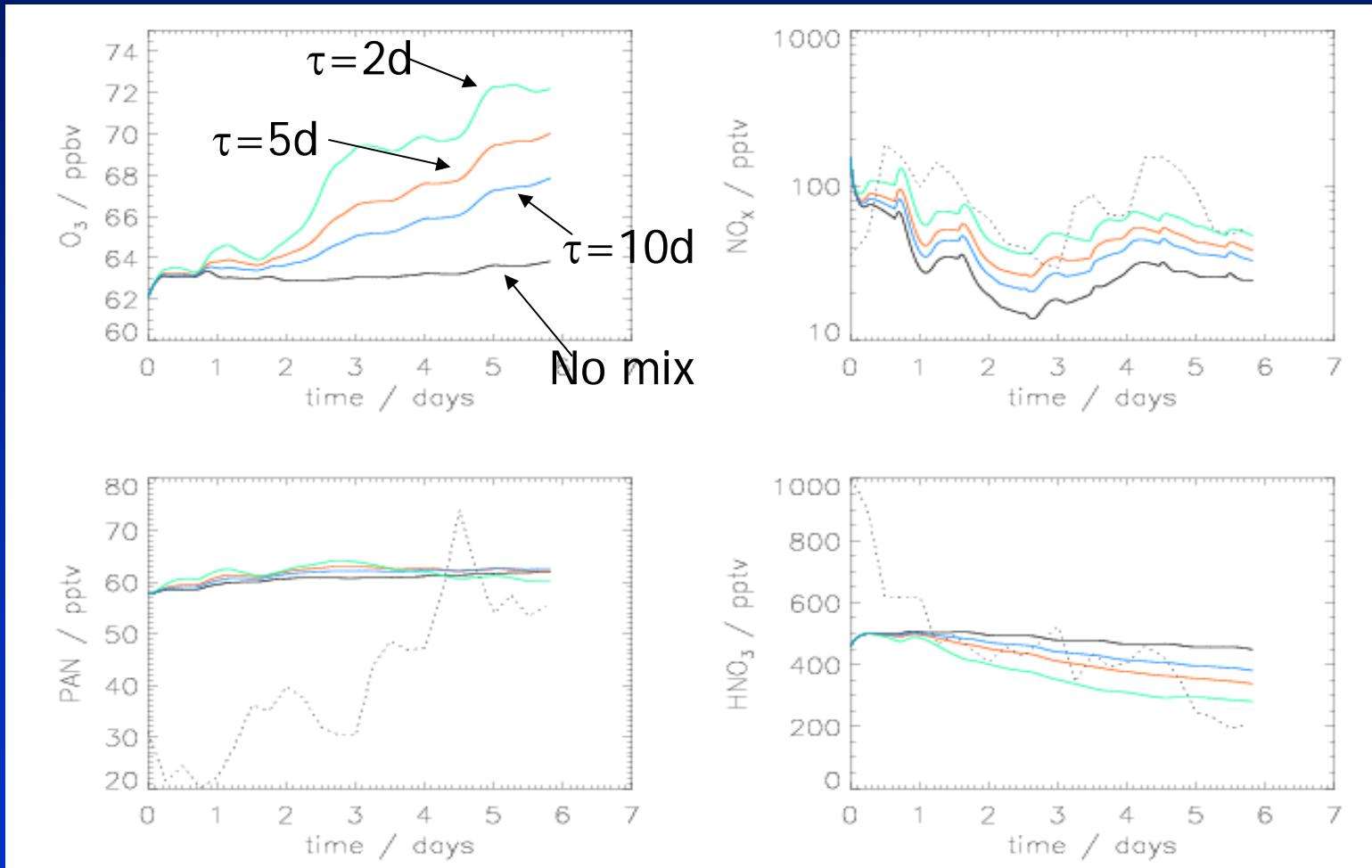
# Sensitivity Analysis

Use Lagrangian model to investigate sensitivity of trans-Atlantic  $\Delta O_3$  to:

- Mixing rate
- Cloud cover
- ECMWF water vapour
- NMHC complexity
- Initial conditions

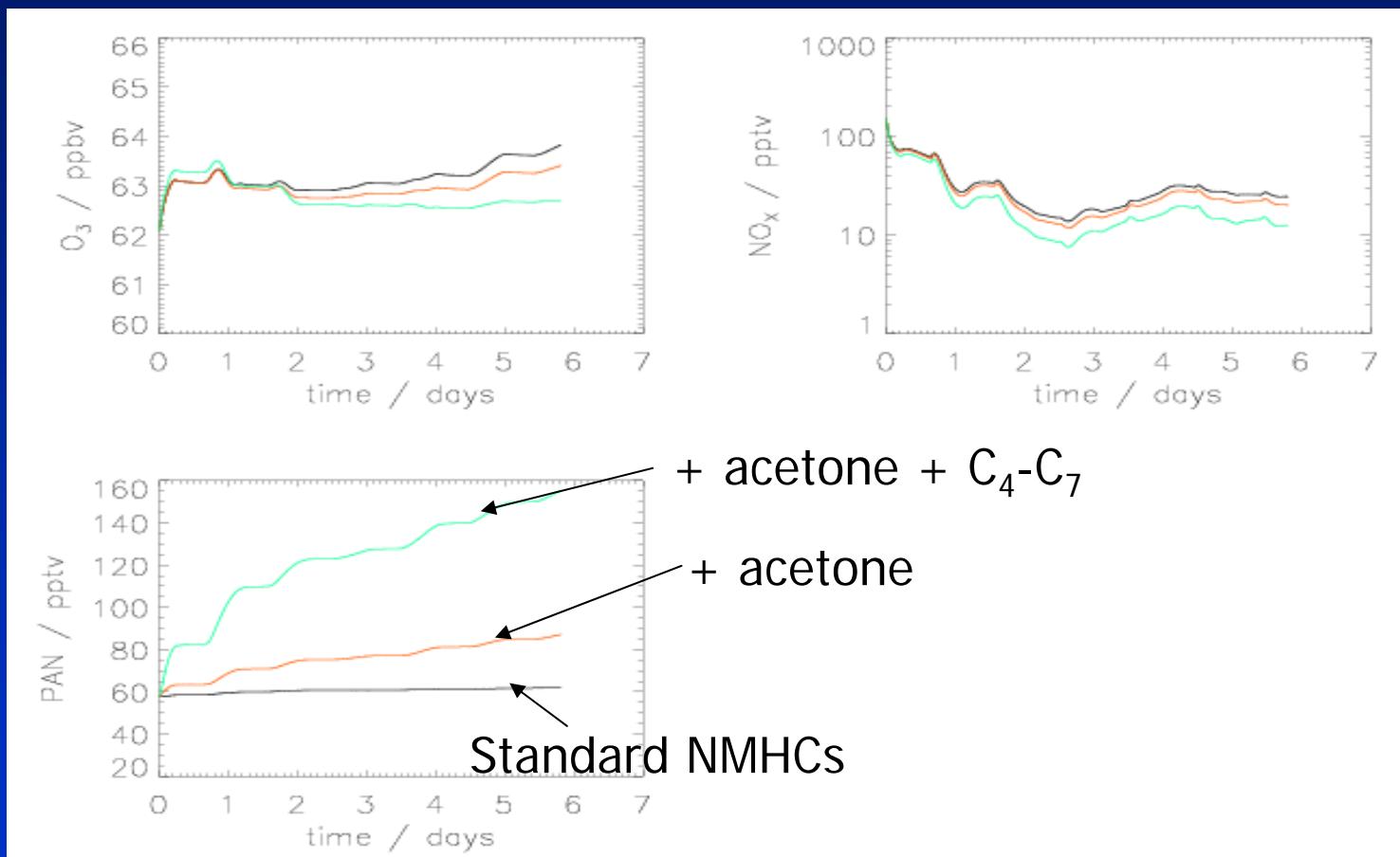


# Mixing Rate



More NOx mixed into air mass  $\Rightarrow$  *more ozone produced*

# NMHC complexity



Formation of PAN through NMHC oxidation  
⇒ *less NO<sub>x</sub> available for ozone production*

# Conclusions

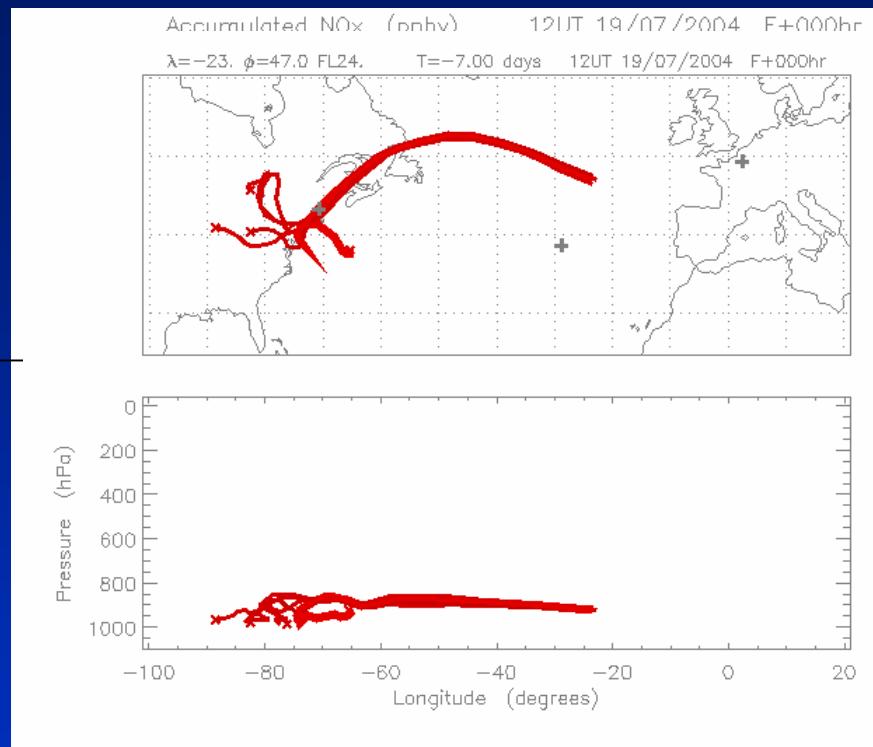
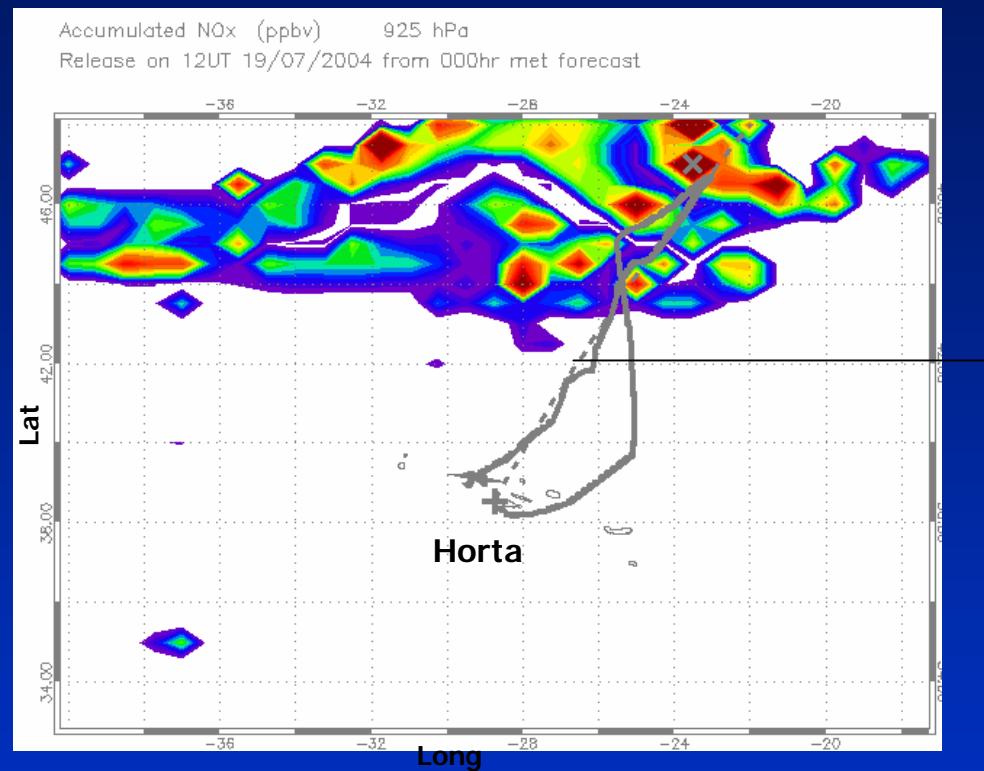
- Trajectory forecasts are sufficiently accurate to execute intercontinental Lagrangian experiment.
- Uncertainties in observed  $\Delta O_3$  and air-mass matches influence diagnosis of trans-Atlantic photochemical  $\Delta O_3$ .

# Investigations Underway

- Constraint of mixing term through hydrocarbons.
- Full Monte-Carlo analysis using Lagrangian model.
- Can reduced chemical mechanisms describe the observed change in composition or is complexity unavoidable?

# Possible multiple interception of New York polluted airmass 2

## First BAE146 interception flight into NYC air 19/7



- NO<sub>x</sub> emissions tracer on 925hPa on 19/7 with BAE146 (air mass relative) flight track overlaid.
- High NO<sub>x</sub> predicted from NYC outflow.
- Obtained using RDF3D trajectories and also accumulating NO<sub>x</sub> from EDGAR emissions inventory when in ECMWF boundary layer.

*Back trajectories calculated from flight track confirm NYC as being at air mass origin*

# Identifying Lagrangian Opportunities

- Selected out of ~30000 trajs from each domain based on criteria:
  1. Passing within range of 2 or 3 bases (*Pease, Azores, Creil*),
  2. Accumulated NOx emissions > threshold (*along back trajs*),
  3. Further NOx emissions < threshold (*along forward trajs*).

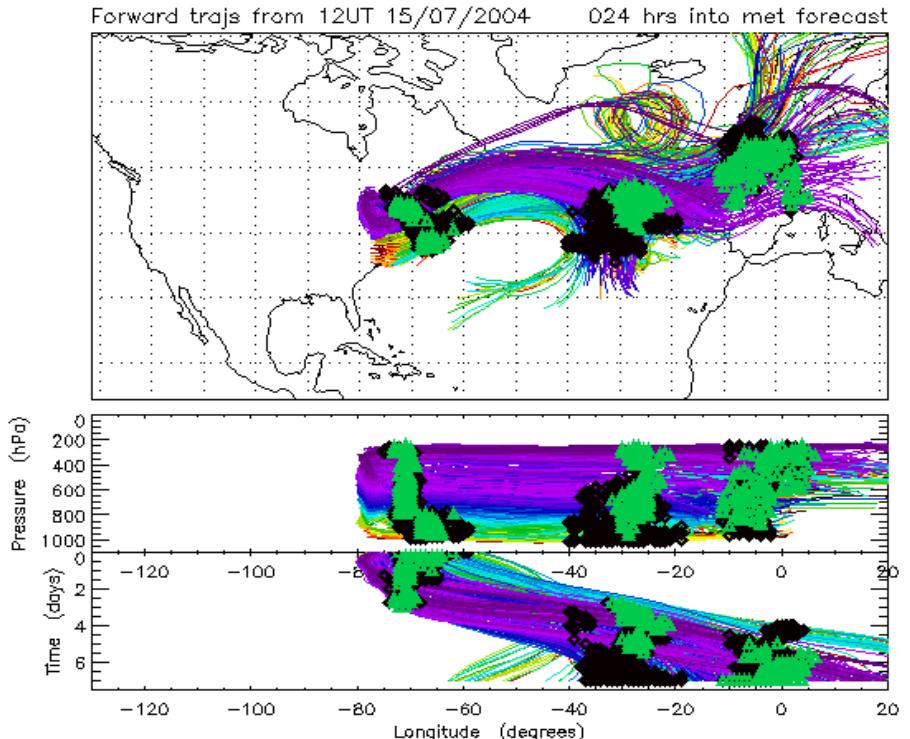
(surface emissions [EDGAR] are picked up within BL as defined by ECMWF forecasts)

## Example

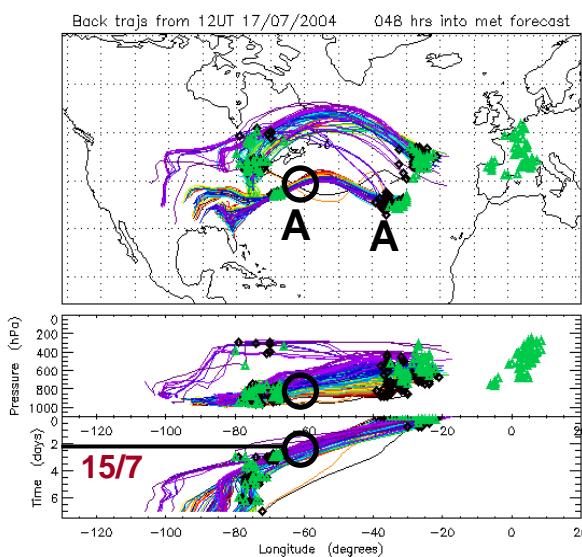
### Forward trajs from US domain

Release time 12UT 15 July 2004  
Based on ECMWF forecast from  
12UT 14 July 2004

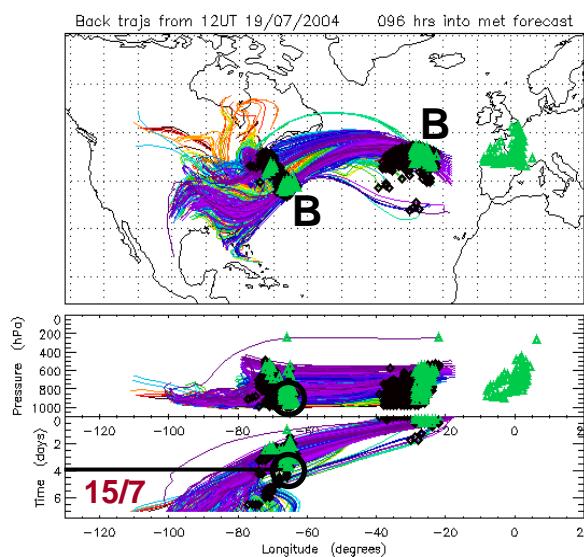
- Black diamonds – 2-point opportunities
- Green triangles – 3-point opportunities (US – Azores – Europe)
- Red squares – 3-point opportunities (any other order)



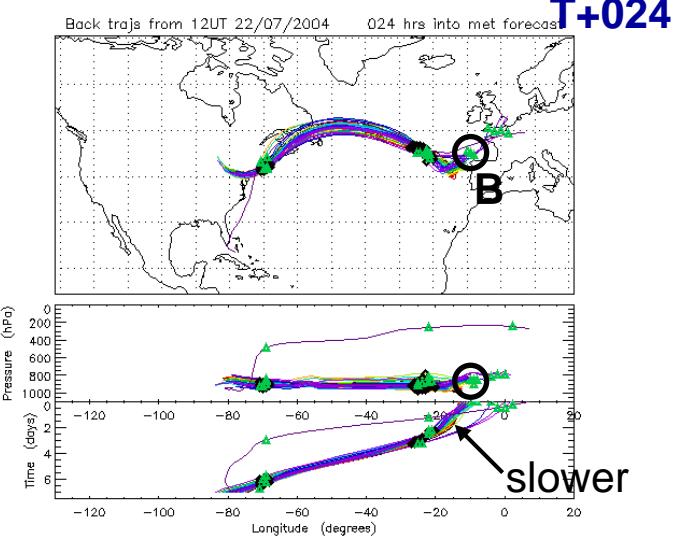
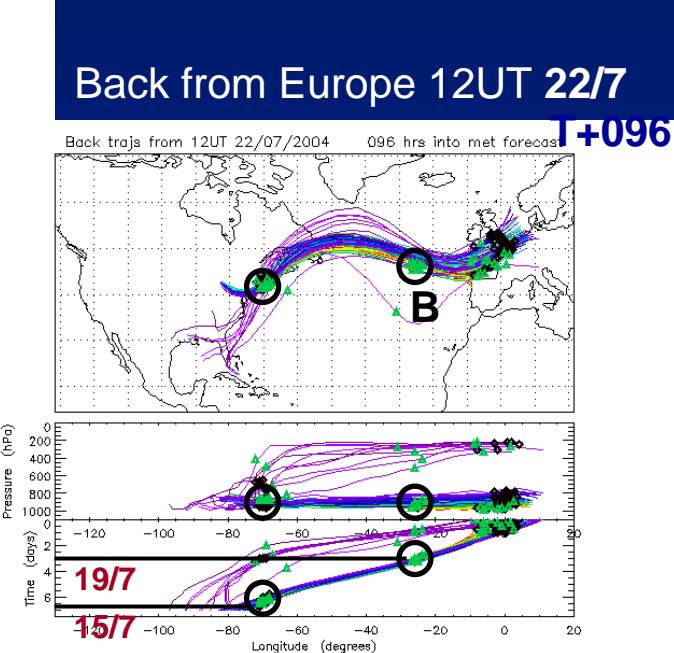
# The first Lagrangian opportunities



Back from Azores 12UT 17/7  
Based on ECMWF forecast  
from 12UT 15/7 (**T+048**)



Back from Azores 12UT 19/7  
Same met forecast (**T+096**)



Note: Back trajectory calculations use wind *analyses* for dates earlier than start of ECMWF forecast.

Note similarity between trajectory forecasts with different lead times (on right). Air masses are predicted to slow down after Azores in later forecasts.

# *Sample collection*

