University of Washington's Activities during INTEX-B:

Results from the Mt. Bachelor Observatory & Duchess aircraft

INTEX-B Data Review Meeting

6 Mar 2007

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along with Prof. Dan Jaffe & Joel Thornton; Duli Chand, Will Hafner, Jeremy Smith; Dylan Millet (Harvard) and Sarah Strode; Louisa Emmons (NCAR)

Questions guiding our research at MBO:

1. What are the inter-annual variations in CO, O_3, PAN, NO_x, NO_y, and aerosols? How do these change in response to large-scale biomass burning, ENSO, variations in transport pathways , etc.?

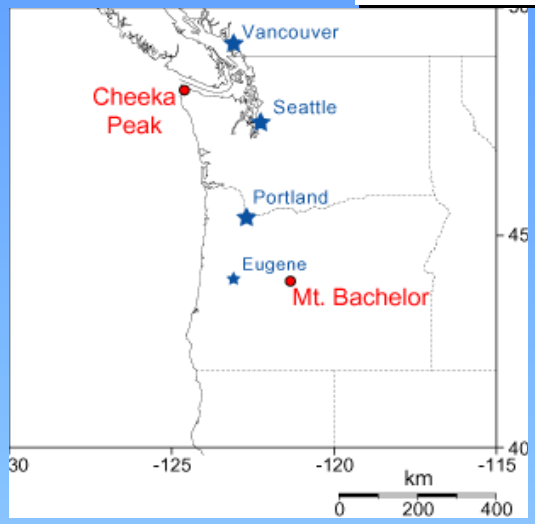
2. How well do satellite observations and global models capture the inter-annual variations in CO, O_3, PAN, NO_y and aerosols? Do variations in PAN correlate with in-situ and satellite observations of tropospheric O_3?

3. How do inter-annual variations in background atmospheric composition influence surface air quality?

4. What gas and aerosol tracer ratios can be used to identify U.S. pollution, Asian industrial pollution, mineral dust, and/or biomass burning at Mt. Bachelor?

5. Can we detect changes in the mixing ratios of CO, PAN and/or O_3 associated with changing upstream (Asian) NO_x emissions? How do these changes impact the oxidative capacity of the troposphere?

Measurement Platforms



DUCHESS AIRCRAFT

Meteorological:

T; RH; P; GPS position

Chemical:

3-λ (450, 550 & 700 nm) σ_{sp} and σ_{bsp}; CO; O_3; Hg^0

MT. BACHELOR OBSERVATORY (MBO)

- Operating continuously since March 2004
- At ~2.7 km ASL, we frequently sample FT air

Meteorological:

T; RH; P; WV; Wind Speed + Direction

Chemical:

σ_{sp}; CO; O_3; Hg^0; PHg; RGM; NO, NO_2; APNs

Publications:

1) Weiss-Penzias et al.: Atmospheric mercury fluxes based on rations with CO in pollution plumes from biomass burning and East Asian pollution sources, *Atmos. Environ.*, 2007
2) Swartzendruber et al.: Observations of reactive gaseous mercury in the free troposphere at MBO, *J. Geophys. Res.*, 2006.
3) Weiss-Penzias et al: Observations of Asian air pollution in the free troposphere at MBO in spring 2004, *J. Geophys. Res.*, 2006
4) Jaffe et al.: Export of atmospheric mercury from Asia, *Atmos. Environ.*, 2005

Main MBO Results from INTEX-B

Mean ± 1 σ	ALL	Wet (80th percentile of WV)	Dry (20th percentile of WV)	Dry - Wet % Difference
H_2O Vapor (g/kg)	2.6 ± 1.0	4.0 ± 0.4	1.2 ± 0.4	-108%
O_3 (ppbv)	49.4 ± 9.7	41.6 ± 8.5	60.4 ± 9.1	38%
CO (ppbv)	142 ± 14	138 ± 16	142 ± 13	3%
RGM (pg/m^3)	31.3 ± 45.4	14.1 ± 18.4	59.3 ± 57.4	144%
PAN (pptv)	339 ± 370	229 ± 186	536 ± 487	91%
PPN (pptv)	23 ± 25	12 ± 10	38 ± 33	113%

Our previous work has found that water vapor is the best indicator of FT air and that most Asian transport events at MBO occur in dry air

GEOS-Chem + MBO during INTEX-B

Full Chemistry

- Pearson correlation coefficients, R
- Mean P at MBO ~734 hPa

	NO_x	O_3	PAN	CO
L3 (~857 hPa)	0.26	0.67	0.29	0.12
L4 (~801 hPa)	0.4	0.71	0.40	0.35
L5 (~731 hPa)	***0.64***	***0.72***	***0.45***	***0.42***
L6 (~650 hPa)	0.55	0.57	0.41	0.28
L7 (~565 hPa)	0.53	0.38	0.23	0.27

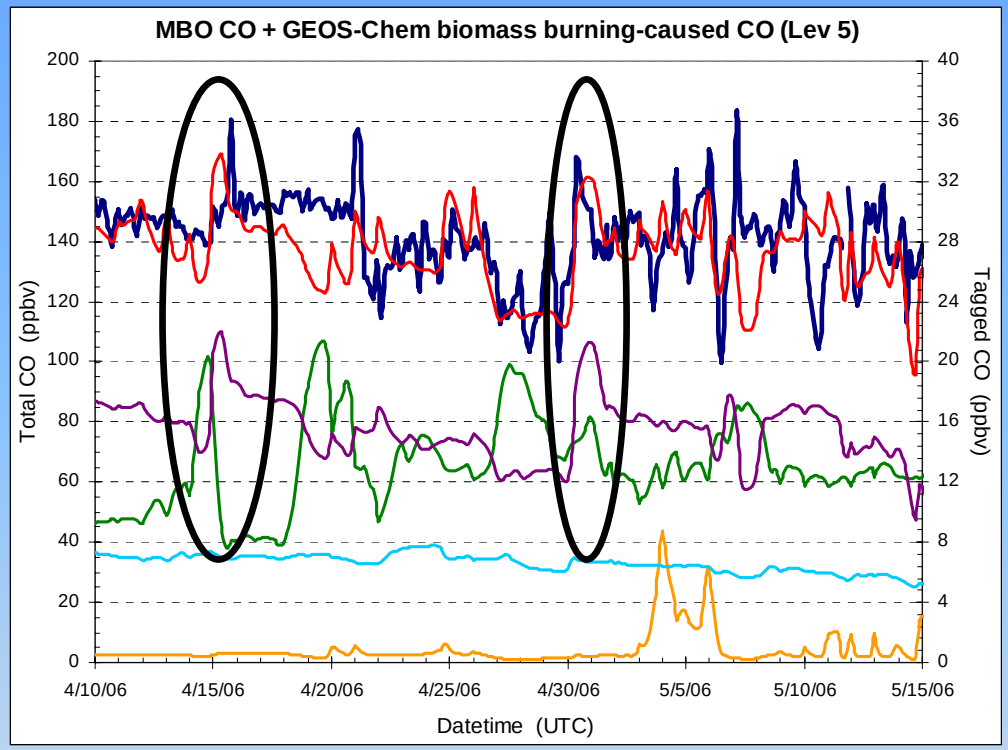
1) Fairly good correlations with NO_x and O_3

2) Qualitatively good with PAN, though large spikes seen in measurements not captured in model

3) CO is systematically lower by ~30 ppbv in model

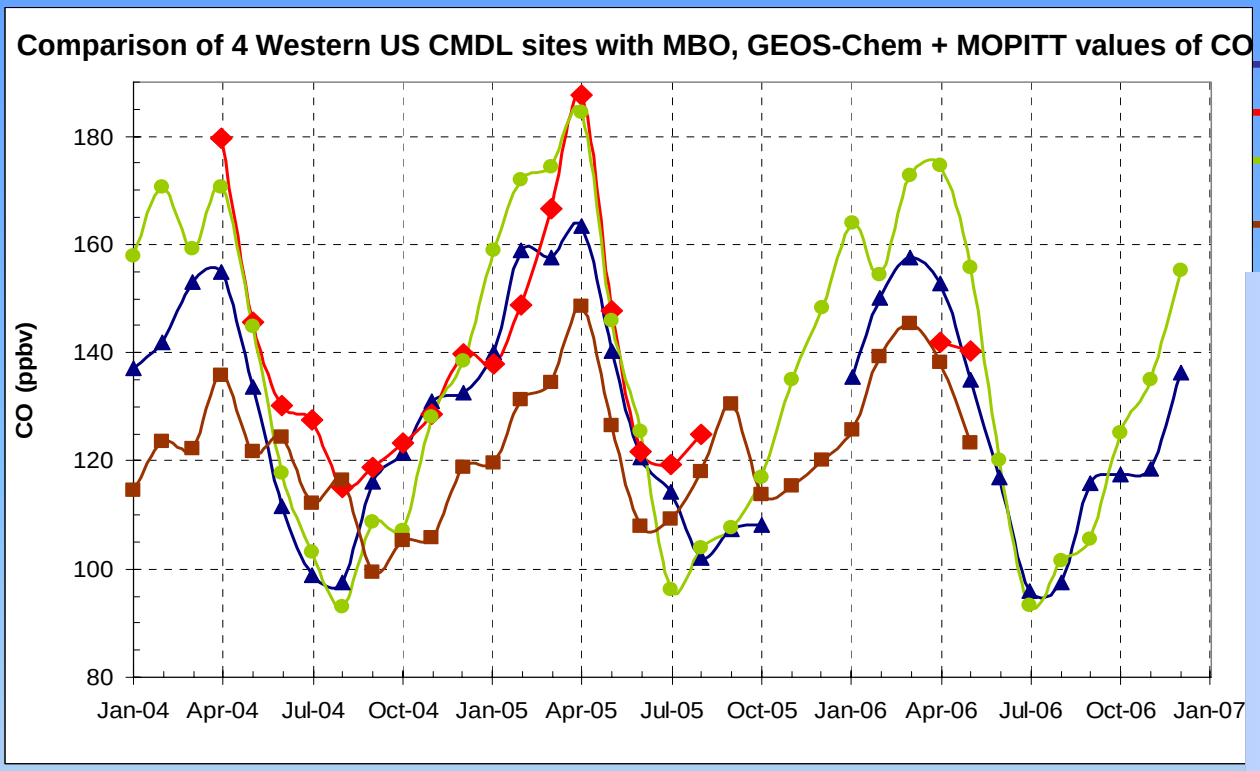
→ MBO vs GEOS-Chem comparisons are best with model Level 5

Tagged CO

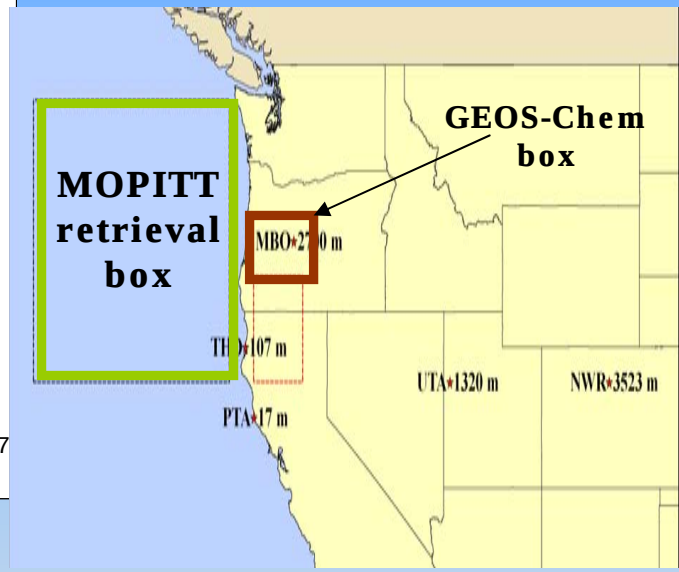


- Total MBO CO
- Total GEOS-Chem CO
- GEOS-Chem CO: N. American biomass burning
- GEOS-Chem CO: Asian biomass burning
- GEOS-Chem CO: Other biomass burning
- GEOS-Chem CO: Global biofuel use

MOPITT, CMDL, GEOS-Chem & MBO CO: Inter-annual variability?



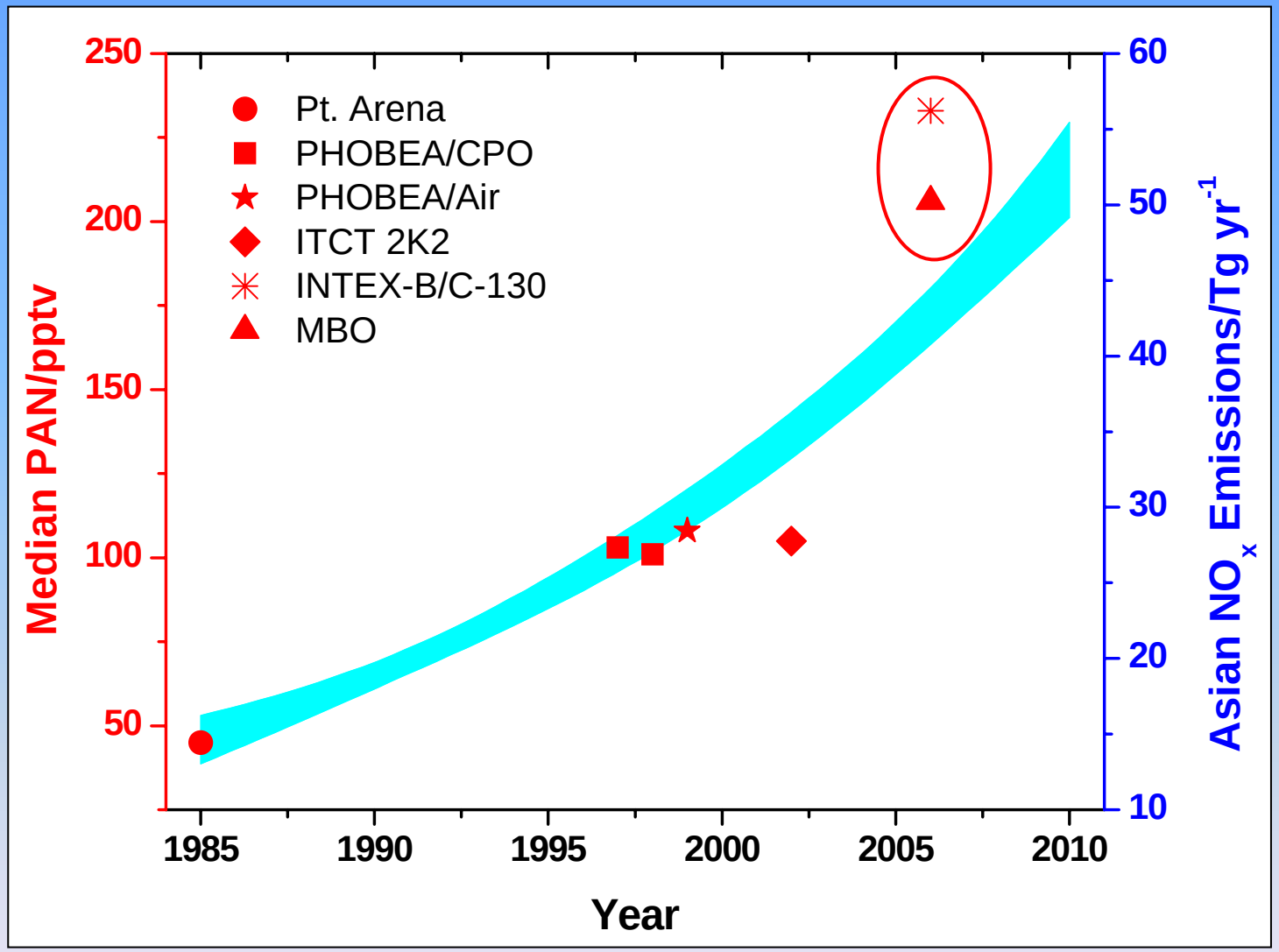
4 CMDL sites
MBO
MOPITT_700 mb
GEOS-Chem



Proposed next steps:

1. Summertime minima are lower in the 700 mb MOPITT retrieval than at MBO probably due to stronger upslope flow. We will test this by comparing the summer FT (i.e., dry) data with MOPITT data
2. Examine inter-annual variability from other available North American West Coast datasets (e.g. Whistler, Cheeka Peak, CMDL sites, etc.)
3. Bring in inter-annual variability in O_3 (OMI?); NO_x (GOME, SCIAMACHY?); other trace gases?
4. Characterize weekly / monthly N. Pacific meteorological patterns (differences in transport pathways)
5. Determine main emission sources (strong fire seasons?; atypically cold winter → more CO?)

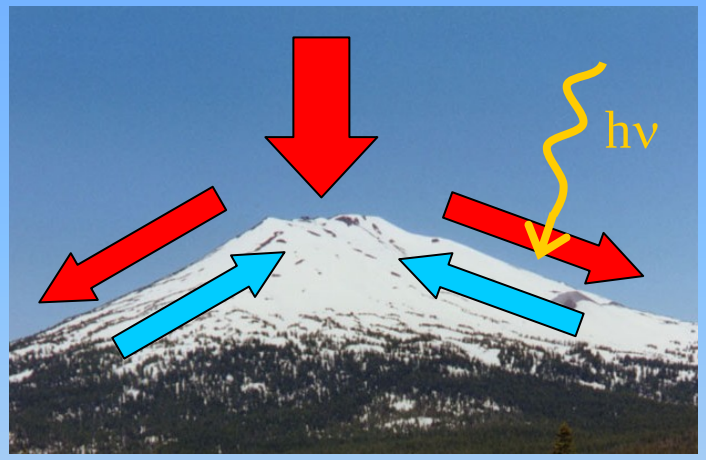
Why We Need More PAN Measurements



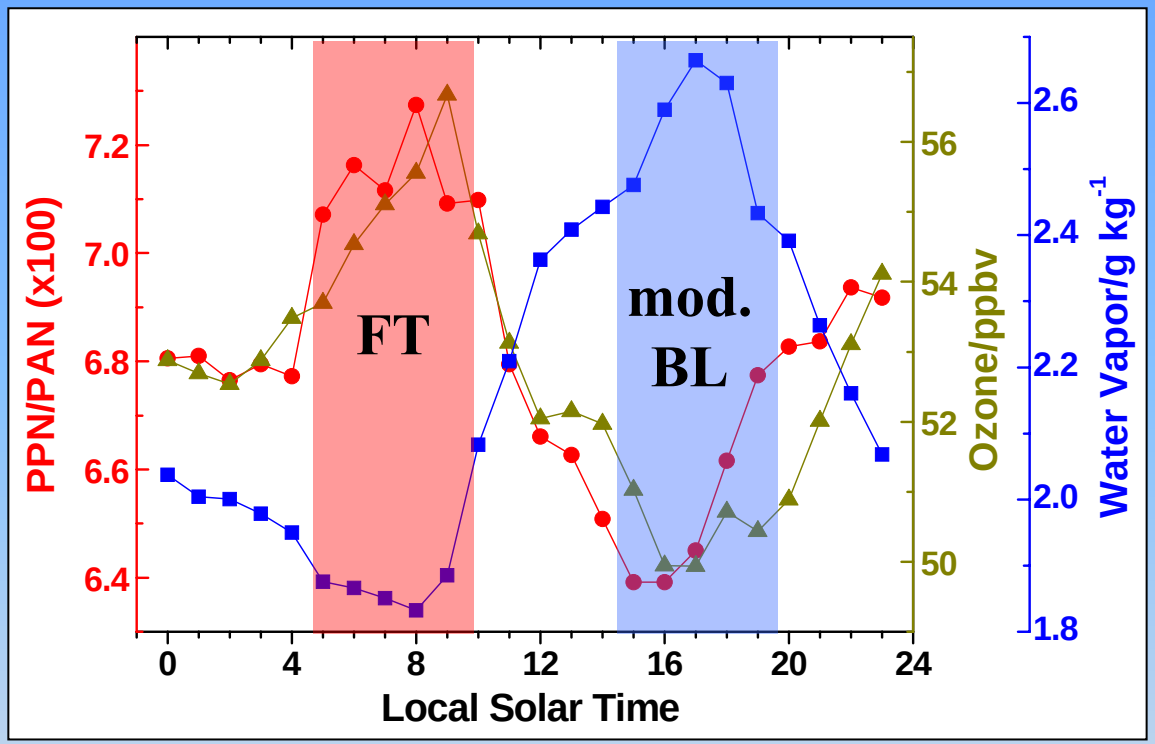
Does NE Pacific PAN mirror Asian NO_x emissions?

Diurnal Behavior

Day: upslope flow brings "modified" BL air



Night: downslope flow and drop in BL height draws FT air

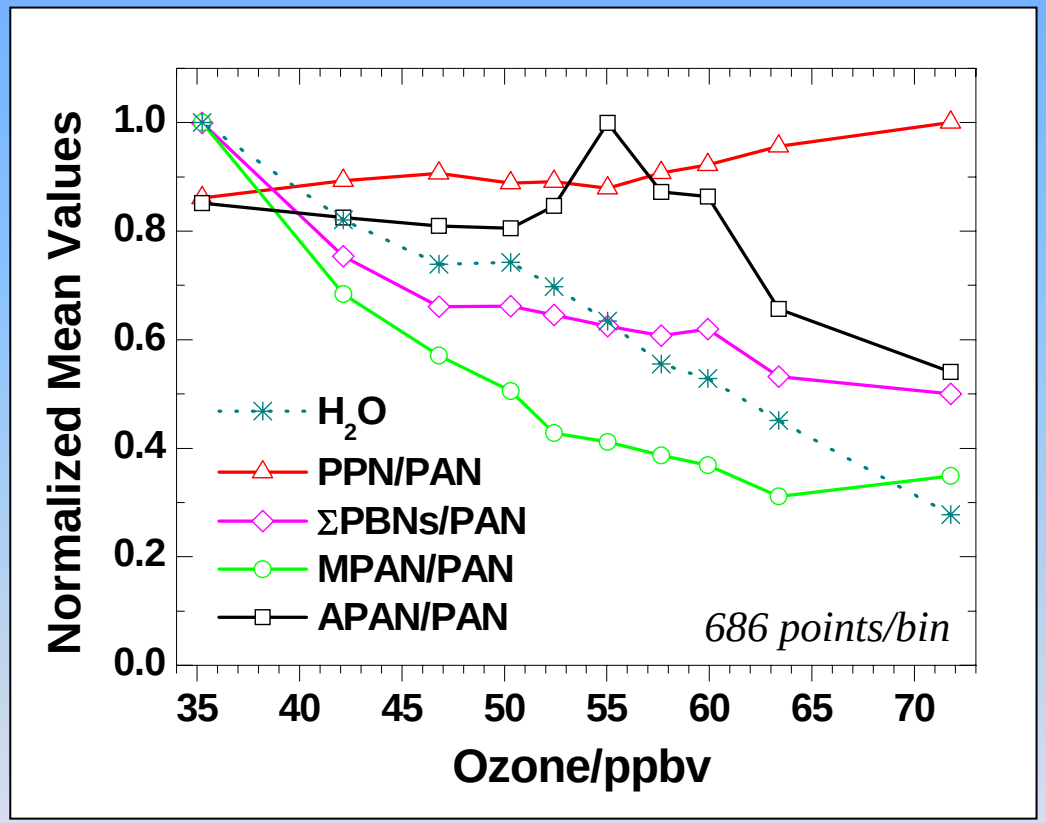


medians	PAN pptv	PPN pptv	PPN/ PAN	O_3 ppbv	NO pptv	Temp °C	H_2O g kg^{-1}	# pts
All Data	206	13	0.068	52.4	4.0	-0.6	2.2	784
Wet	162	9	0.056	42.5	4.2	2.7	3.3	163
Dry	379	27	0.074	64.5	0.4	-2.0	1.0	170

Relationship of O_3 with APN/PAN Ratios

BL ⟷ **FT**

- **Lower O_3**
- **Higher ratios for longer-chain APNs**



- **Higher O_3**
- **Lower ratios for longer-chain APNs**

Highest values of ozone and PAN are associated with *non-local* FT air

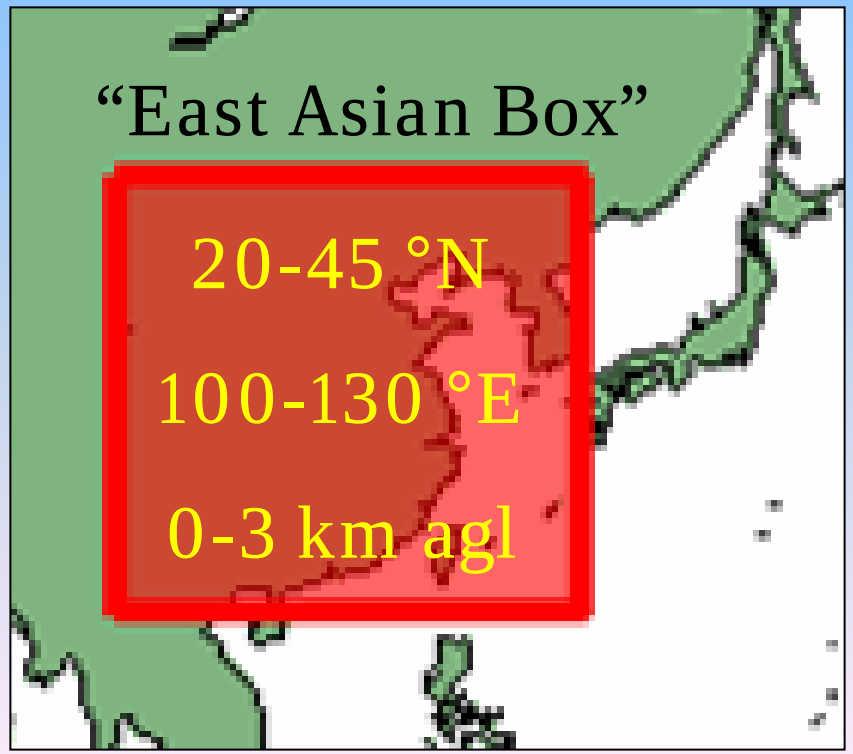
Diagnosing Long-Range Transport

HYSPLIT 10-day Back Trajectories

- GDAS Meteorology
- 30 start alts from 0 – 2.9 km
- 25 lat/lon pairs per altitude
- 784 start times

Total = 588,000 trajectories

Count # of hours in the East Asian box for back trajectories passing over MBO



The Contribution of Pollution from Asia

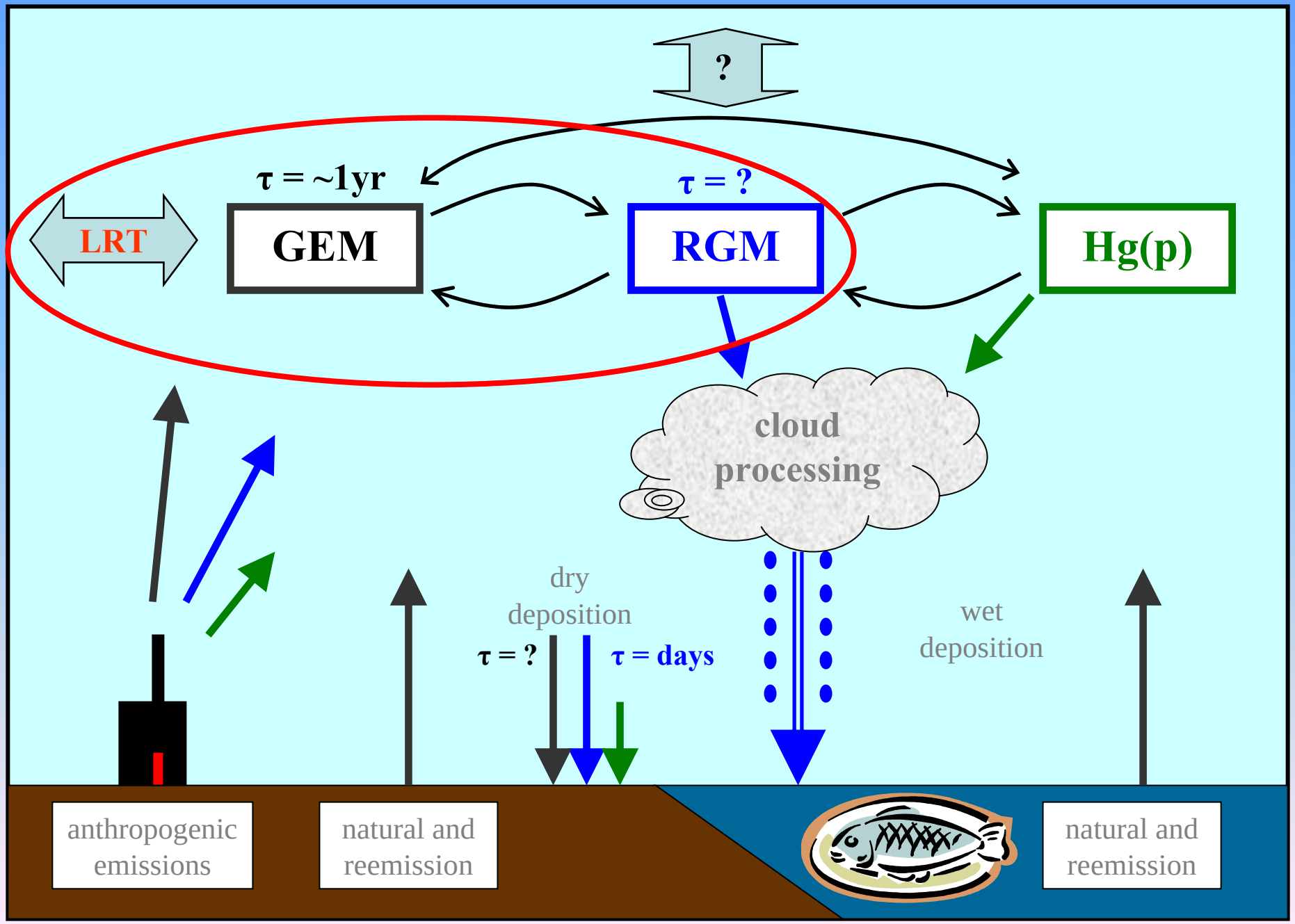
ALRT Index = mean of "hours in the box" for back trajectories with initialization heights from 1.7 – 2.1 km

medians	PAN pptv	PPN pptv	PPN/ PAN	O_3 ppbv	H_2O g kg^{-1}	# pts
ALL DATA	206	13	0.068	52.4	2.2	784
ALRT = 0	148	10	0.065	49.0	2.5	338
ALRT > 0	284	19	0.069	55.3	2.0	446
ALRT + DRY	414	29	0.074	64.5	1.0	126

Also, **$\Delta(Hg^0)/\Delta(CO)$ = 0.0049 ng m^{-3} $ppbv^{-1}$** for ALRT air, consistent with observations in individual Asian plumes (*Jaffe et al. 2005*)

What are the implications of LRT of PANs for air quality in the Pacific Northwest?

Vertical Distribution of Gaseous Elemental Mercury in the Pacific Northwest During the Spring of 2006


?
τ = ~1yr
τ = ?
LRT
GEM
RGM
Hg(p)
cloud processing
dry deposition
τ = ?
τ = days
wet deposition
anthropogenic emissions
natural and reemission
natural and reemission

Prior Work

Asian Long-Range Transport

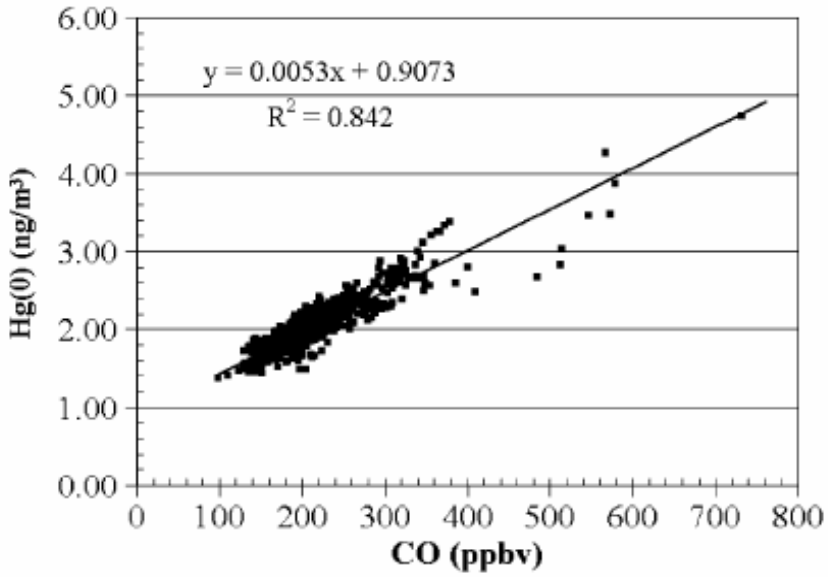
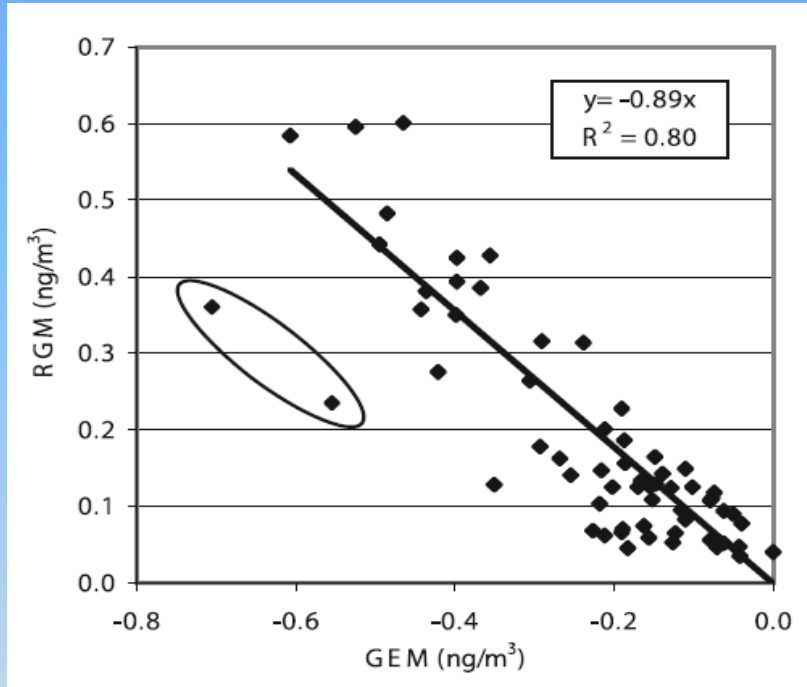


Fig. 2. Scatterplot of Hg^0 vs. CO for all Okinawa data.

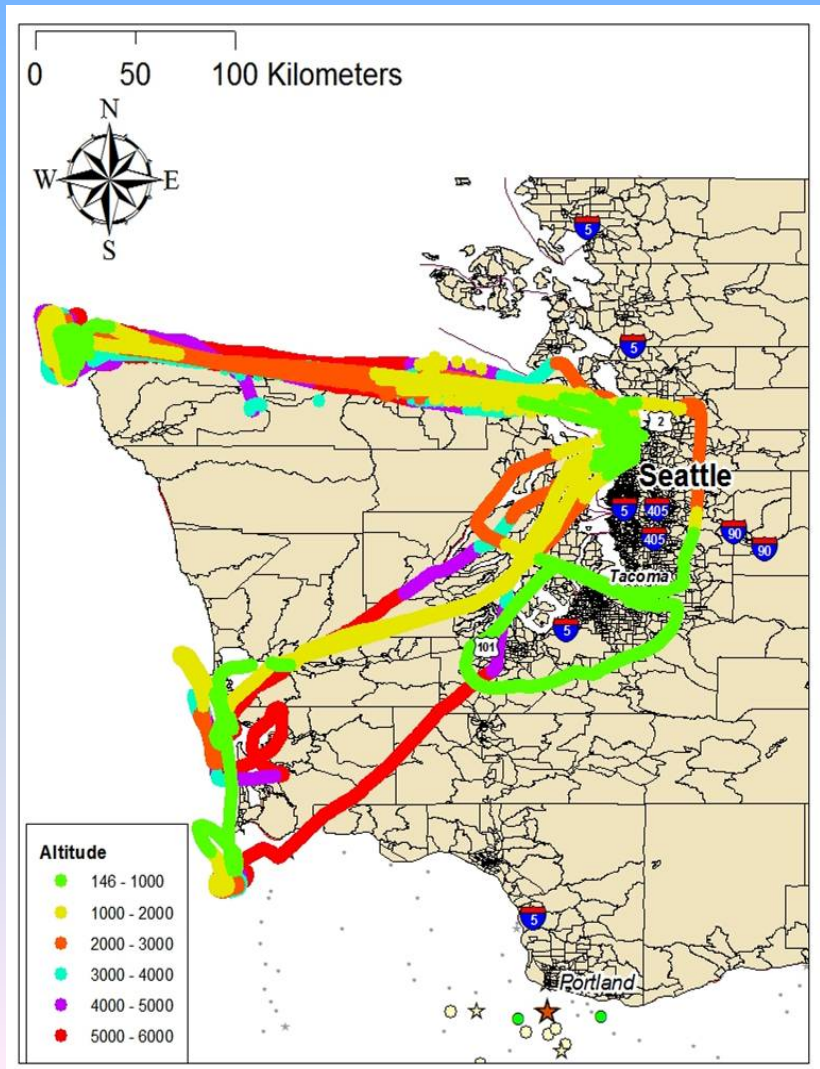
- Jaffe et al (2005) showed strong Hg-CO correlation as indicator of LRT of Asian industrial plume.
- Confirmed by Weiss et al. (2006)

Free Tropospheric Speciation



- Swartzendruber et al (2006) showed inverse relationship between RGM and GEM with sum of species conserved. This implies *in situ* conversion of species

Beechcraft Duchess, 8 flights in Western WA

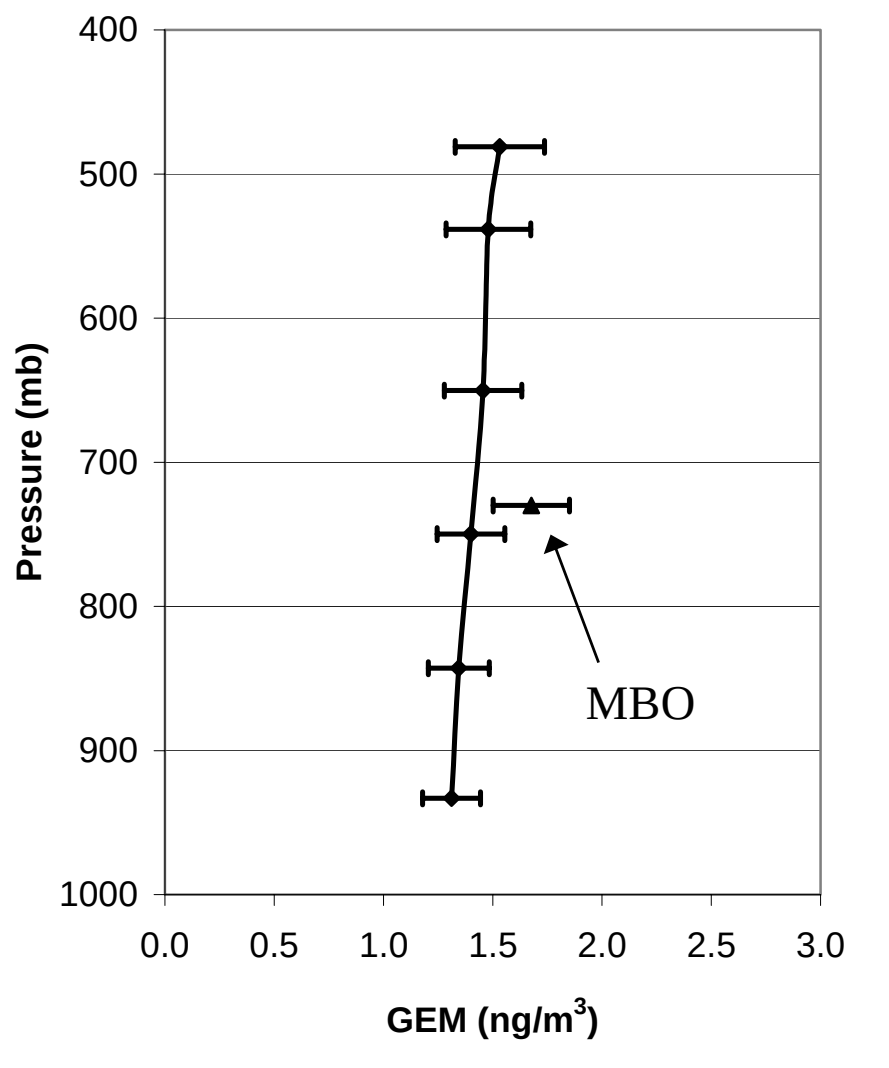


Observations:

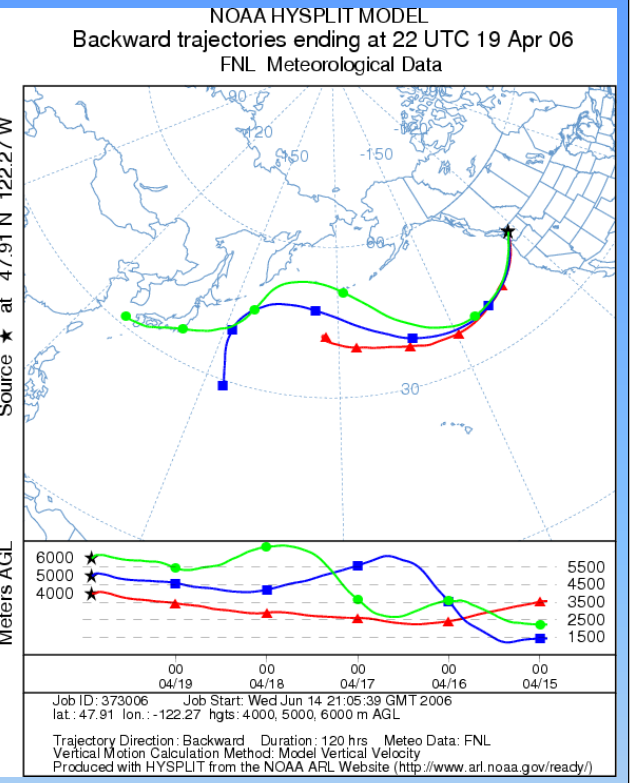
- ozone
- CO
- GEM (Tekran 2537A)
- aerosol scatter
- temperature, RH, GPS

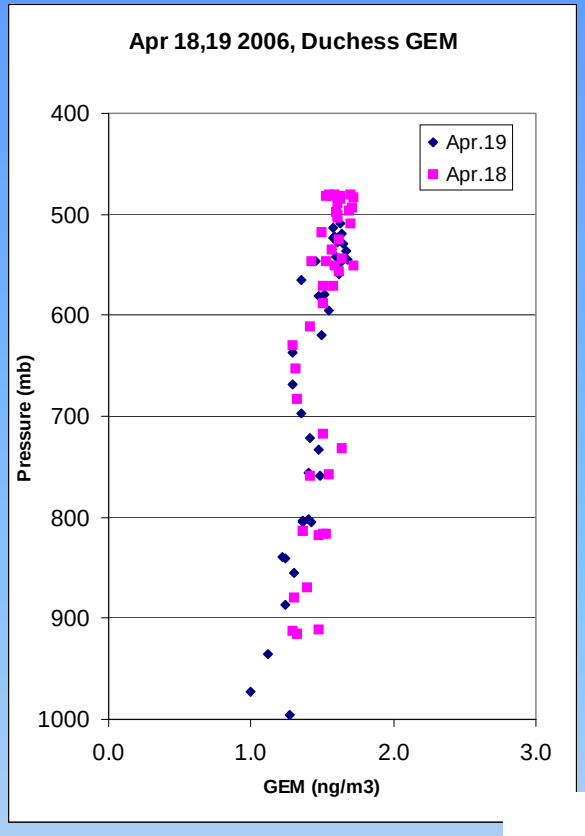
Results: Mean Mercury Profile

- Use KCl scrubber to define method as Gaseous Elemental Mercury (Hg^0)
- Flights chosen to focus on LRT of Asian industrial plume
- Mean profile of all flights
- With GEM from MBO
- Error bars are uncertainty



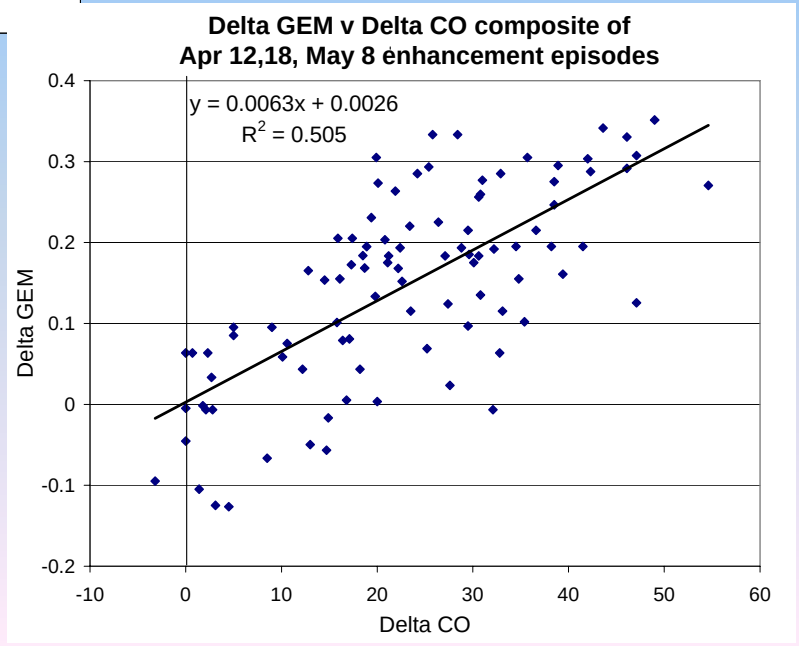
Asian LRT





- Apr 18,19 event shows good precision
- back trajectory to eastern industrial Asia

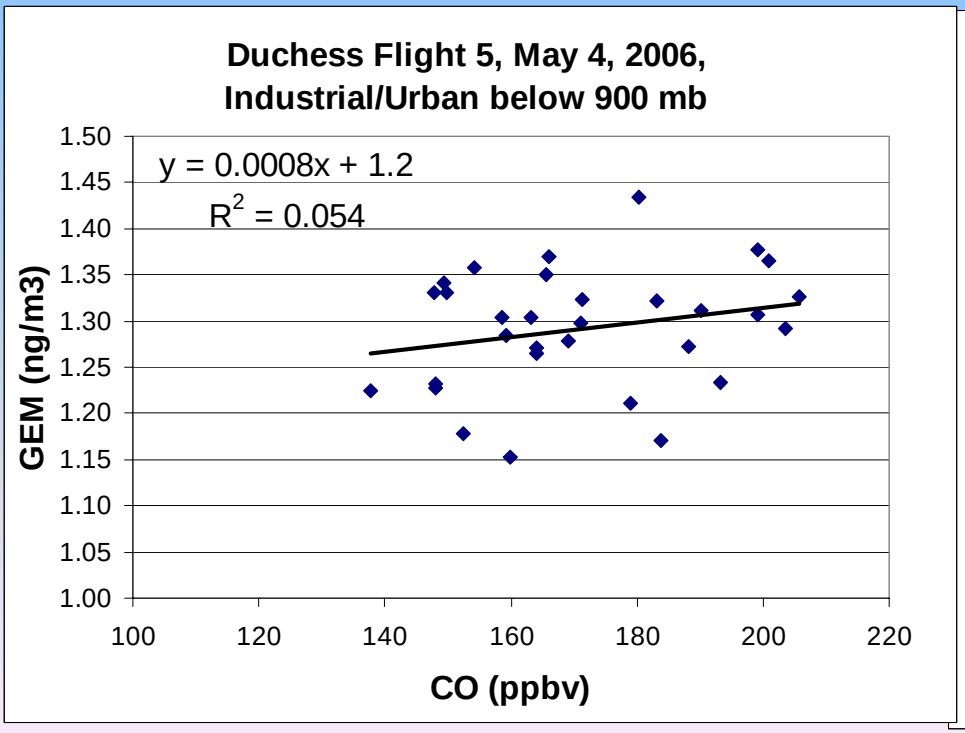
- GEM-CO (ER) of 0.0063 ng/m^3/ppbv
- compares well with Weiss et al. (2006) 0.003 – 0.007 at MBO (2004,5)
- and Jaffe et al. (2005) report 0.004 – 0.007 at Okinawa, JP (2004)

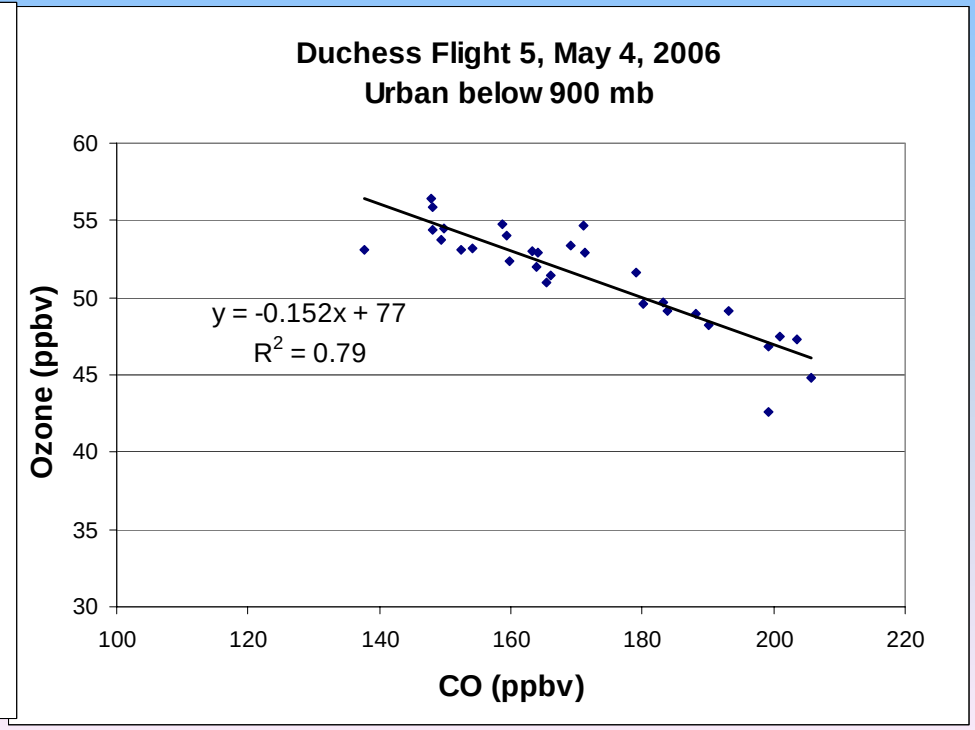


Contrast with Local Flight

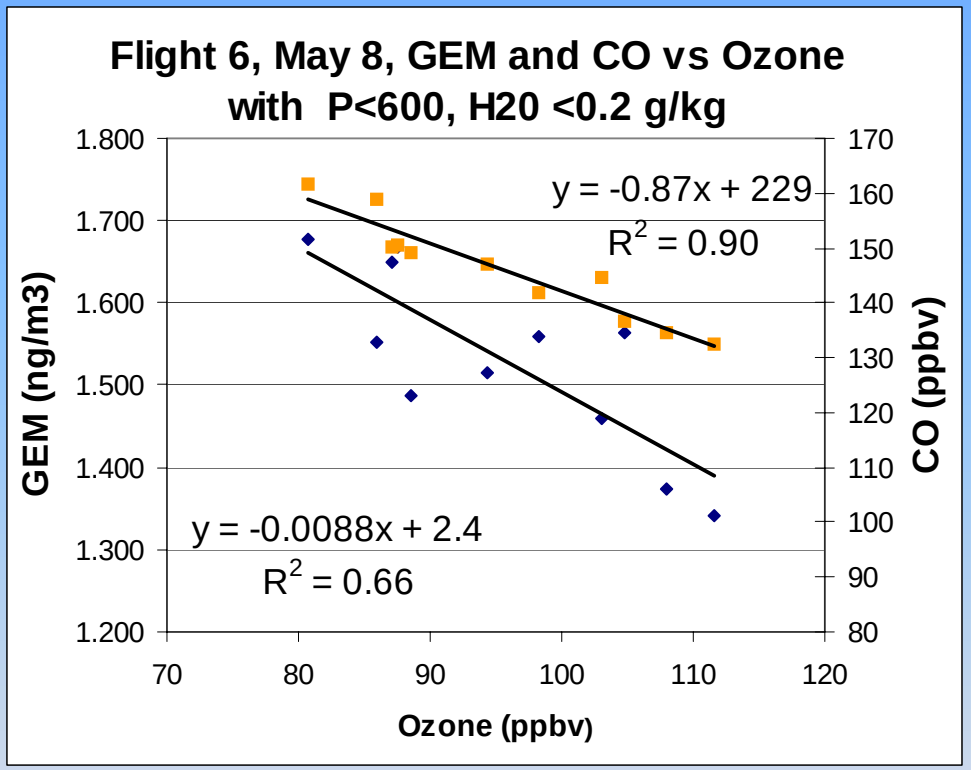
	GEM	*CO*	*Ozone*	*TSG*
CO	0.23	1		
Ozone	-0.25	**-0.89**	1	
TSG	0.09	**0.76**	**-0.64**	1
H20	-0.02	**0.71**	**-0.72**	**0.73**

- no significant GEM-CO enhancement or correlation
- strong anthropogenic tracers in ozone loss environment

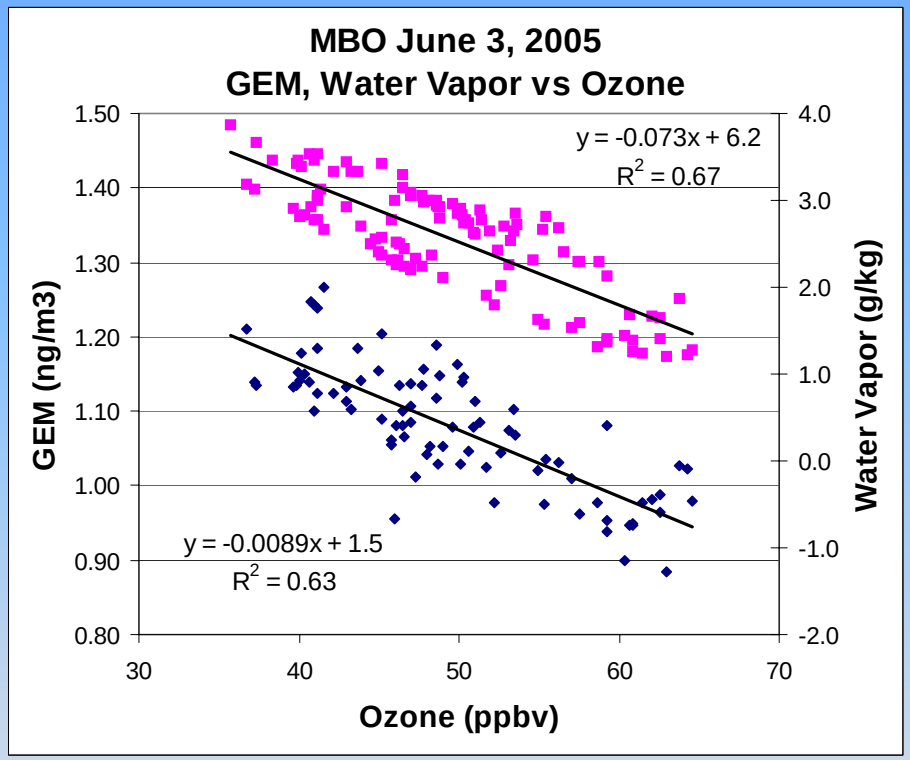




GEM depletion signature suggests Stratospheric/Upper Trop. RGM



Duchess data



- MBO 2005 data
- here, high time resolution shows same signature as RGM rich air described by Swartzendruber et al. (2006)

Acknowledgements

- EPA STAR Grant, NASA and NSF
- Mt Bachelor, Inc
- Carol Higgenbothen, Central Oregon Comm. Coll.
- Staci Simonich Group, Oregon State

MBO and Duchess Aircraft Posters

Dave Reidmiller *et al.*: Results from the Mt. Bachelor Observatory: Our role in INTEX-B and comparisons to MOPITT satellite retrievals of CO and GEOS-Chem model results

Glenn Wolfe *et al.*: Acyl peroxy nitrates in the remote Pacific Northwest: Comparison with previous measurements and diagnosing the influence of long-range pollution transport

Phil Swartzendruber *et al.*: Hg as a tracer of Asian air and FT oxidation of Hg