Global atmospheric distributions of some short-lived halocarbons

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NSF-sponsored HIPPO team
AGAGE team

Focus today on:

	Nominal "Lifetime" (days)**	Loss process
CH_2Br_2	123 d	OH predominantly
$CHBr_3$	24 d	mostly photolysis
CH_3I	7 d	photolysis predominantly

These gases:

→ may account for ~25% of total bromine reaching the stratosphere
→ influence tropospheric oxidation processes (ozone; Hg deposition)
→ have natural sources predominantly

But,

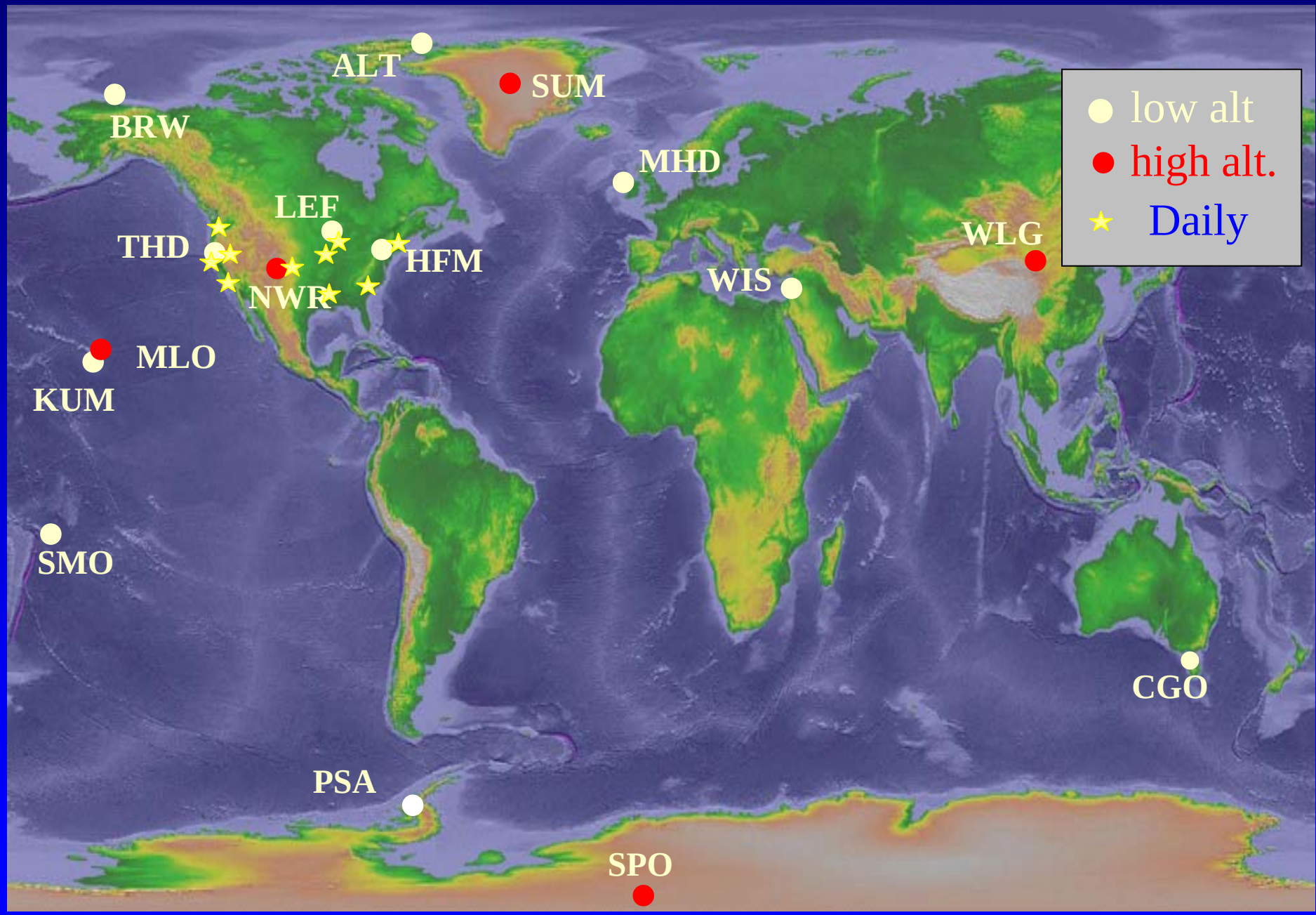
these influences are not well quantified in part because of our poor understanding of their sources and atmospheric distributions (loss rates are comparable to transport times).

Underlying question:

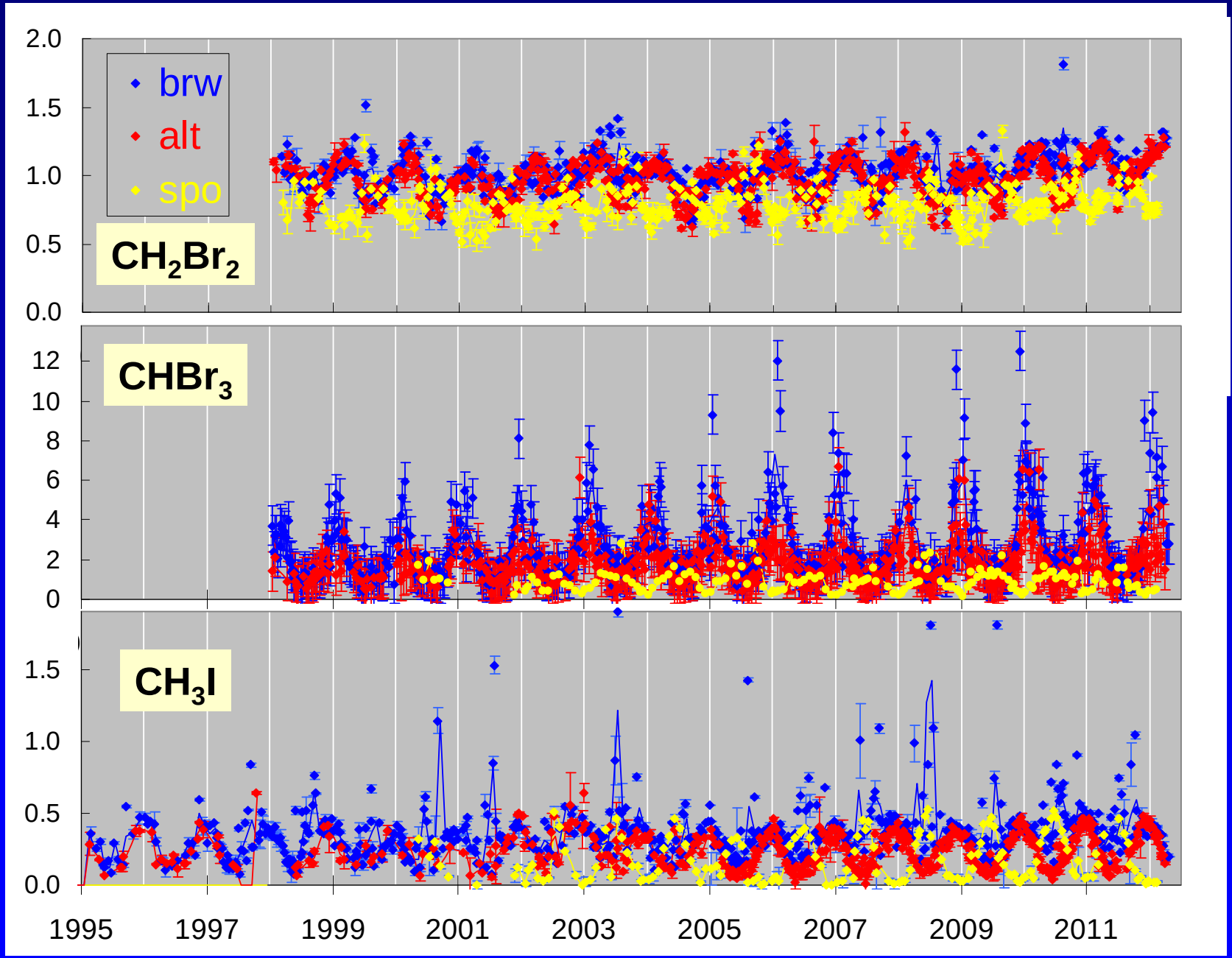
To what extent can "background" atmospheric distributions of short-lived gases be defined?

** **nominal lifetimes** calculated for OH = 1e6 rad cm^{-3} and photolysis at 5 km (WMO, 2003 and 2011)

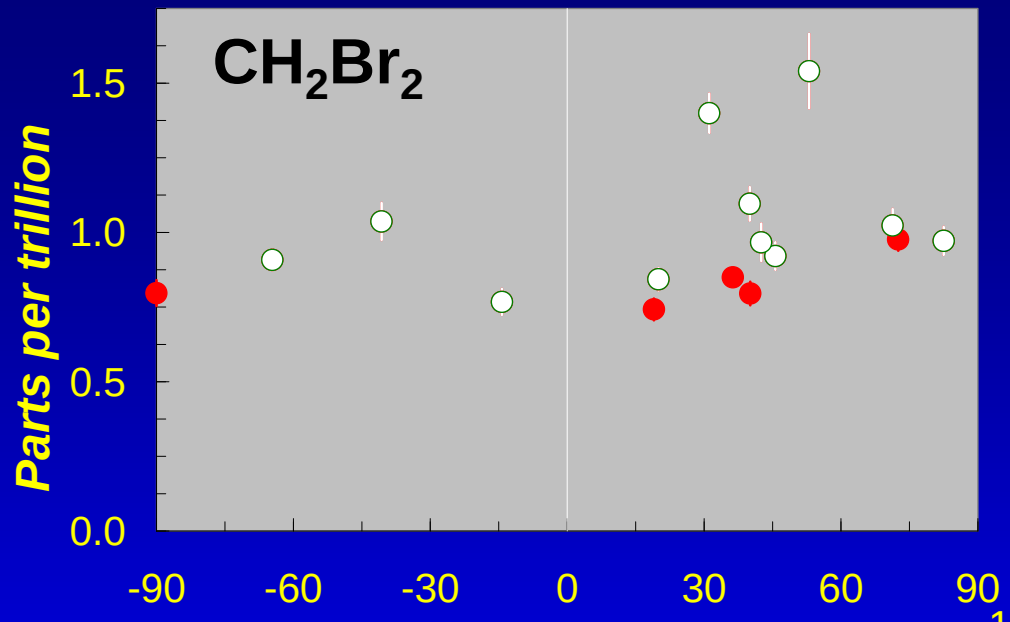
Halocarbon Surface Sampling Network ~weekly flasks



Surface flask data from select sites (pair mean and s.d.)



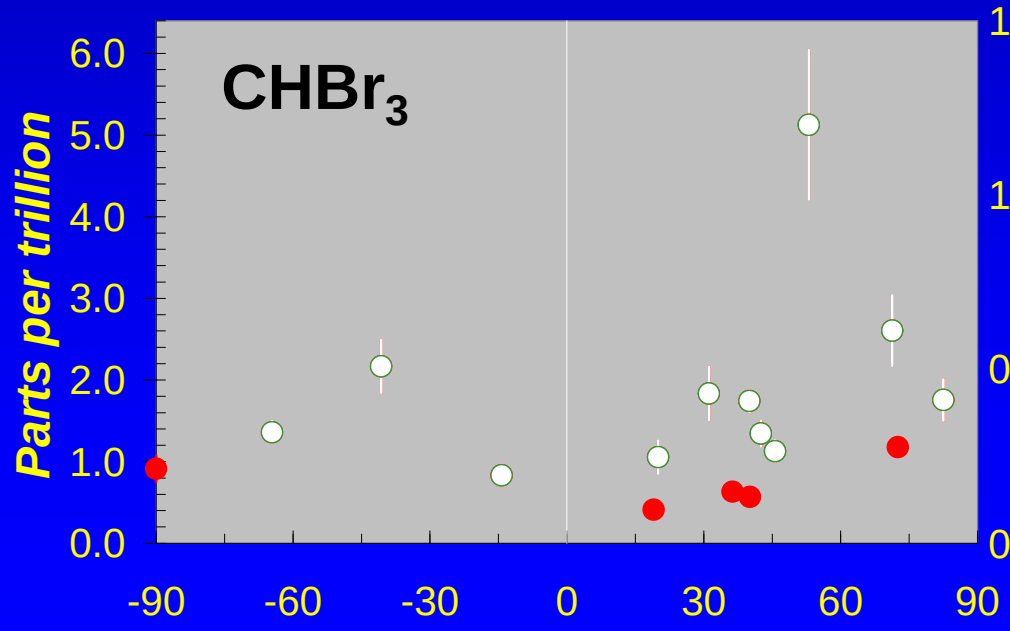
Annual means at surface sites *(2 to 17 yr records; ~wkly. sampling)*

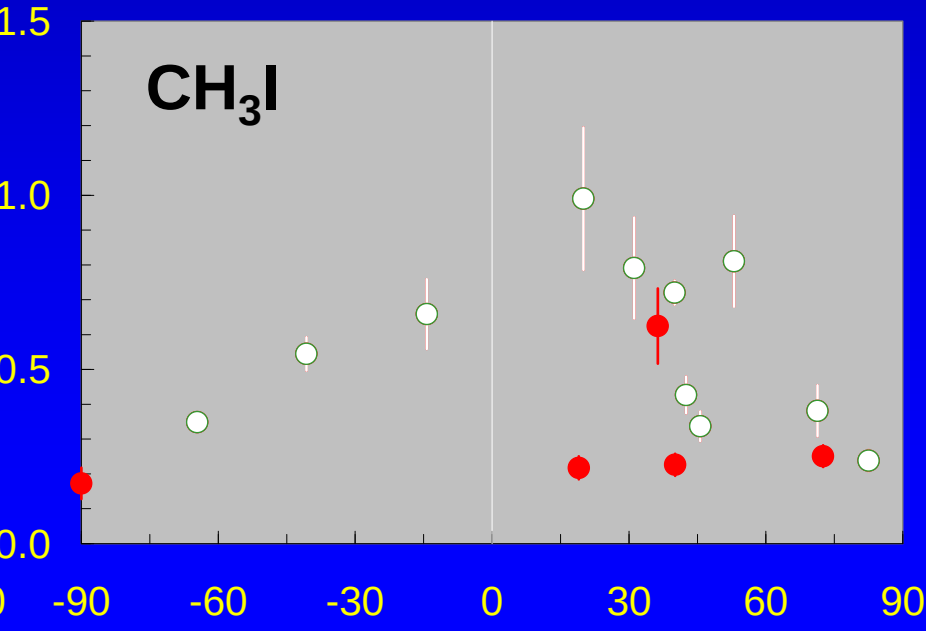


Low altitude sites
High altitude sites, ≥2.8 km asl

Error bars = 1 s.d. of yearly means

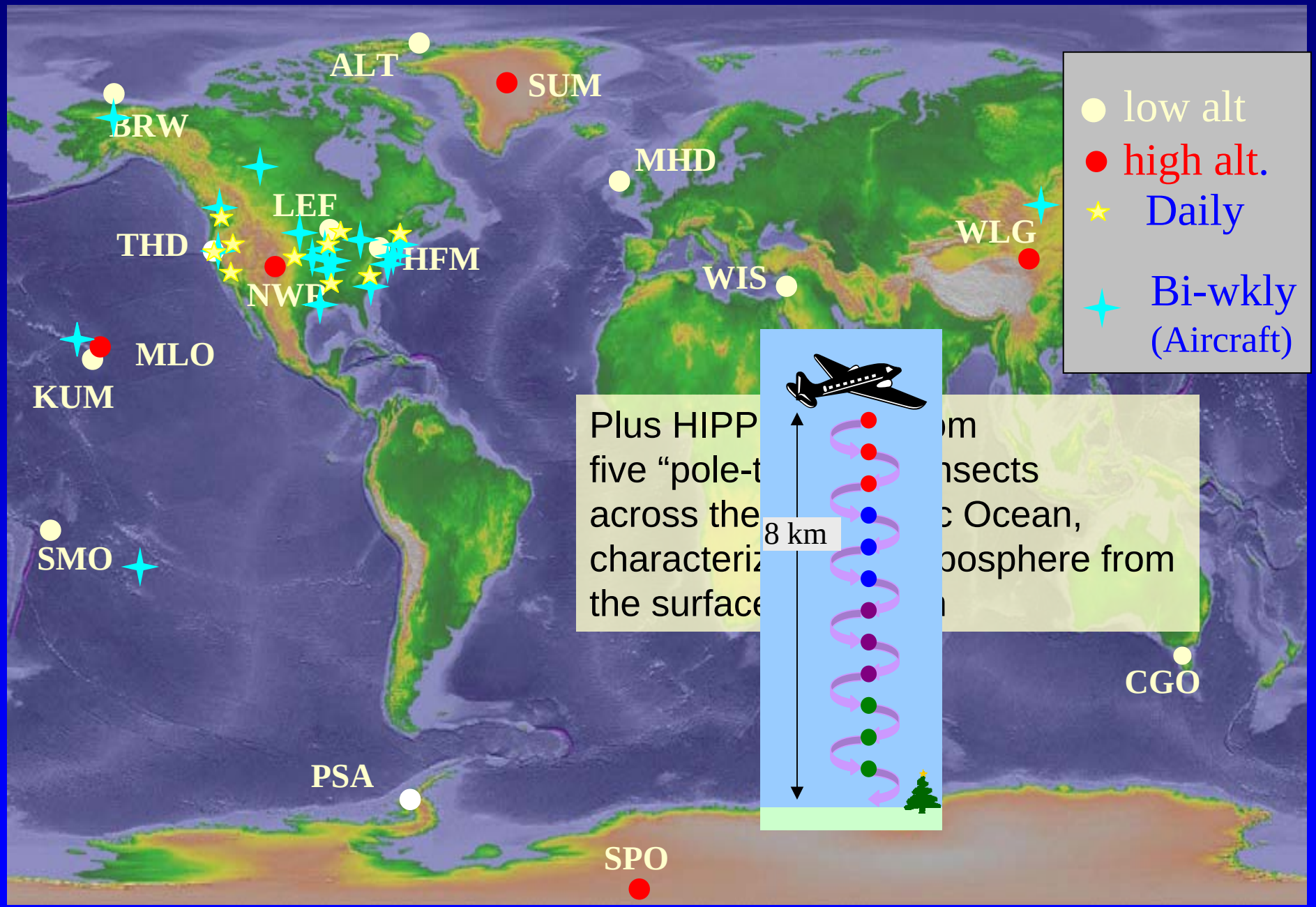
→ Small inter-annual variability
→ Large site-to-site differences
...except at high altitudes





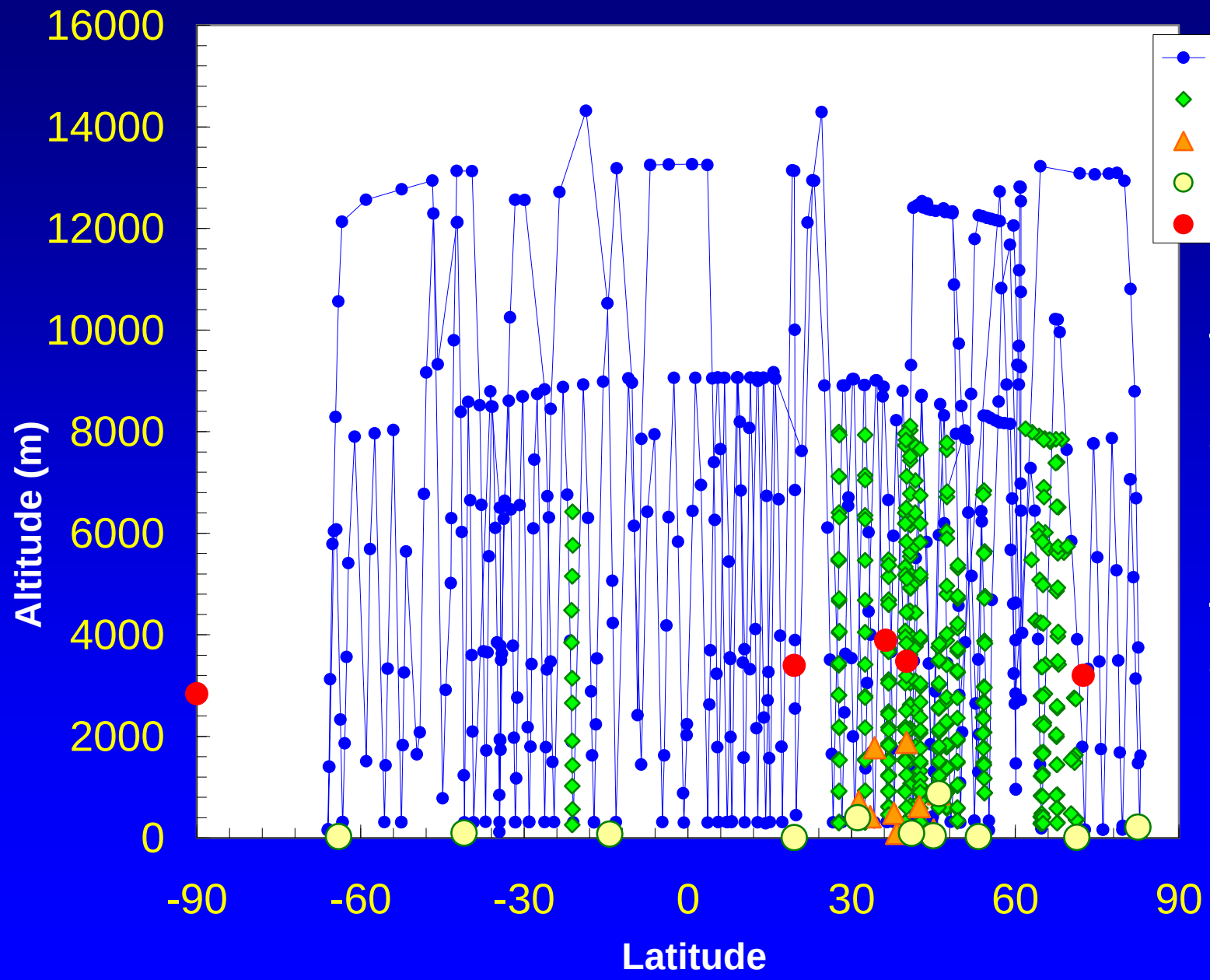
WLG=2; WIS=4; SUM=6; THD=9 yrs

Halocarbon Surface and Aircraft Sampling Network



Sample locations during November 2009…

the 2nd HIPPO deployment

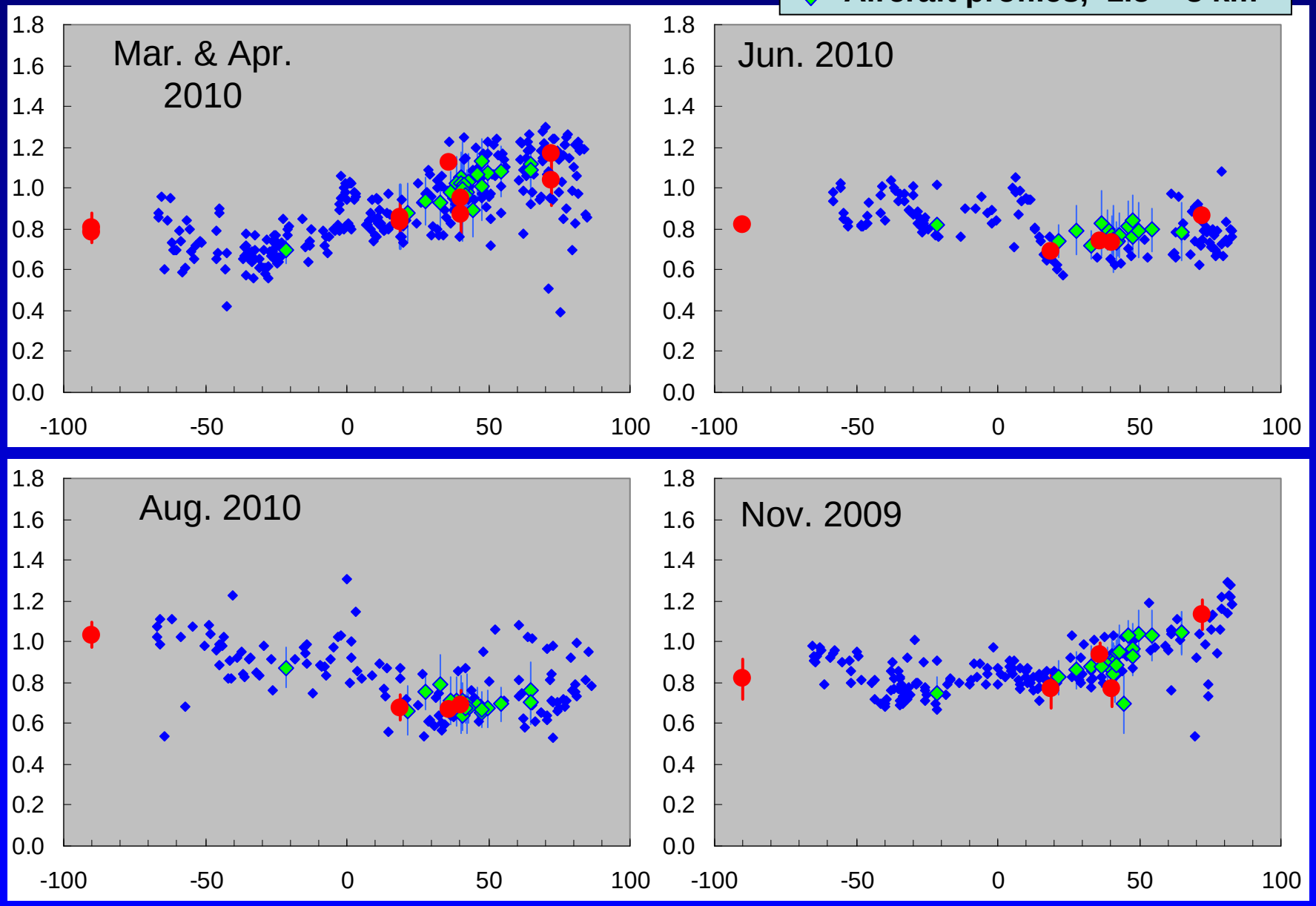


NOAA aircraft samples are predominantly from over North America

HIPPO samples taken from the Pacific Ocean basin

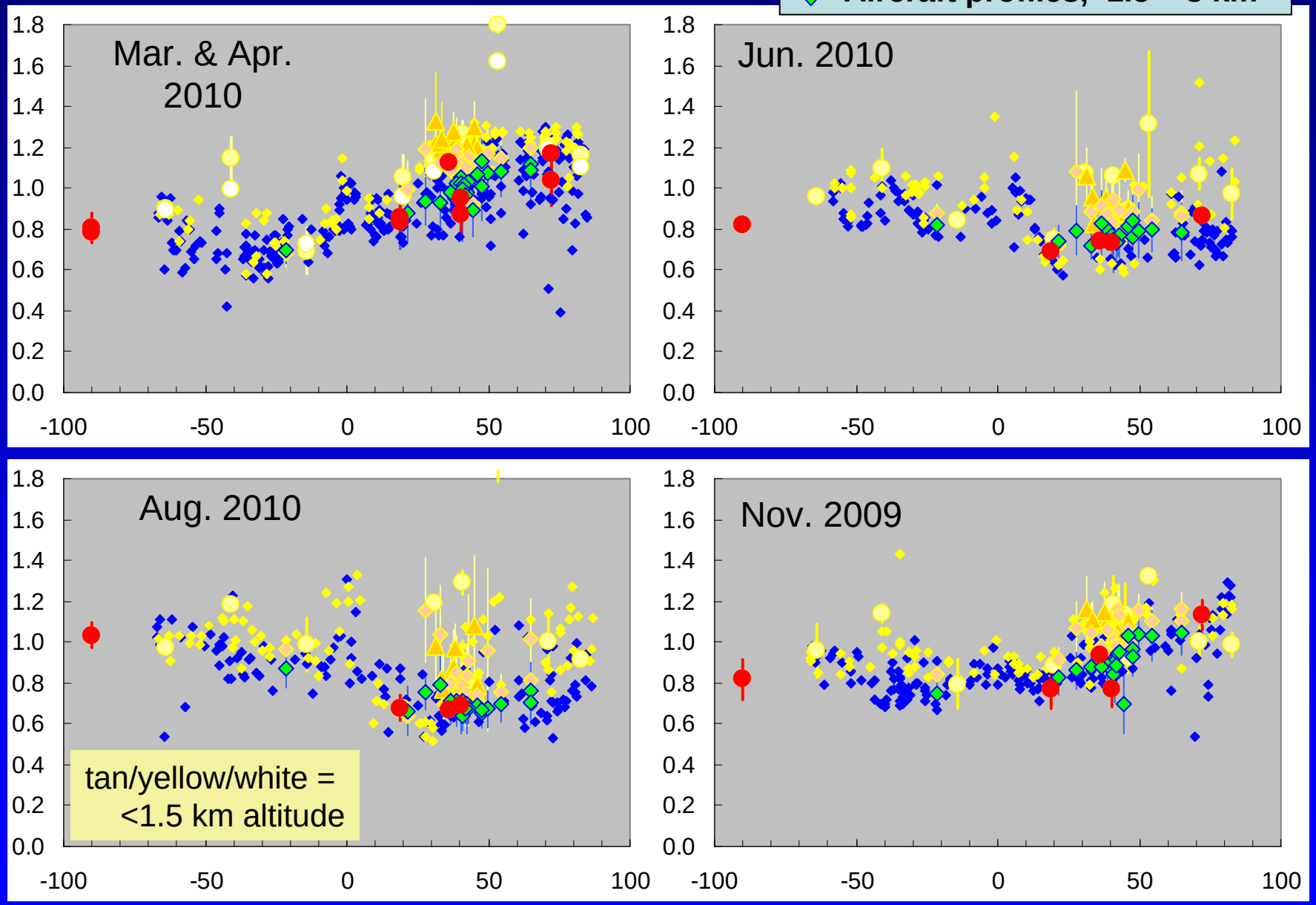
CH_2Br_2 in the FREE troposphere

in different months



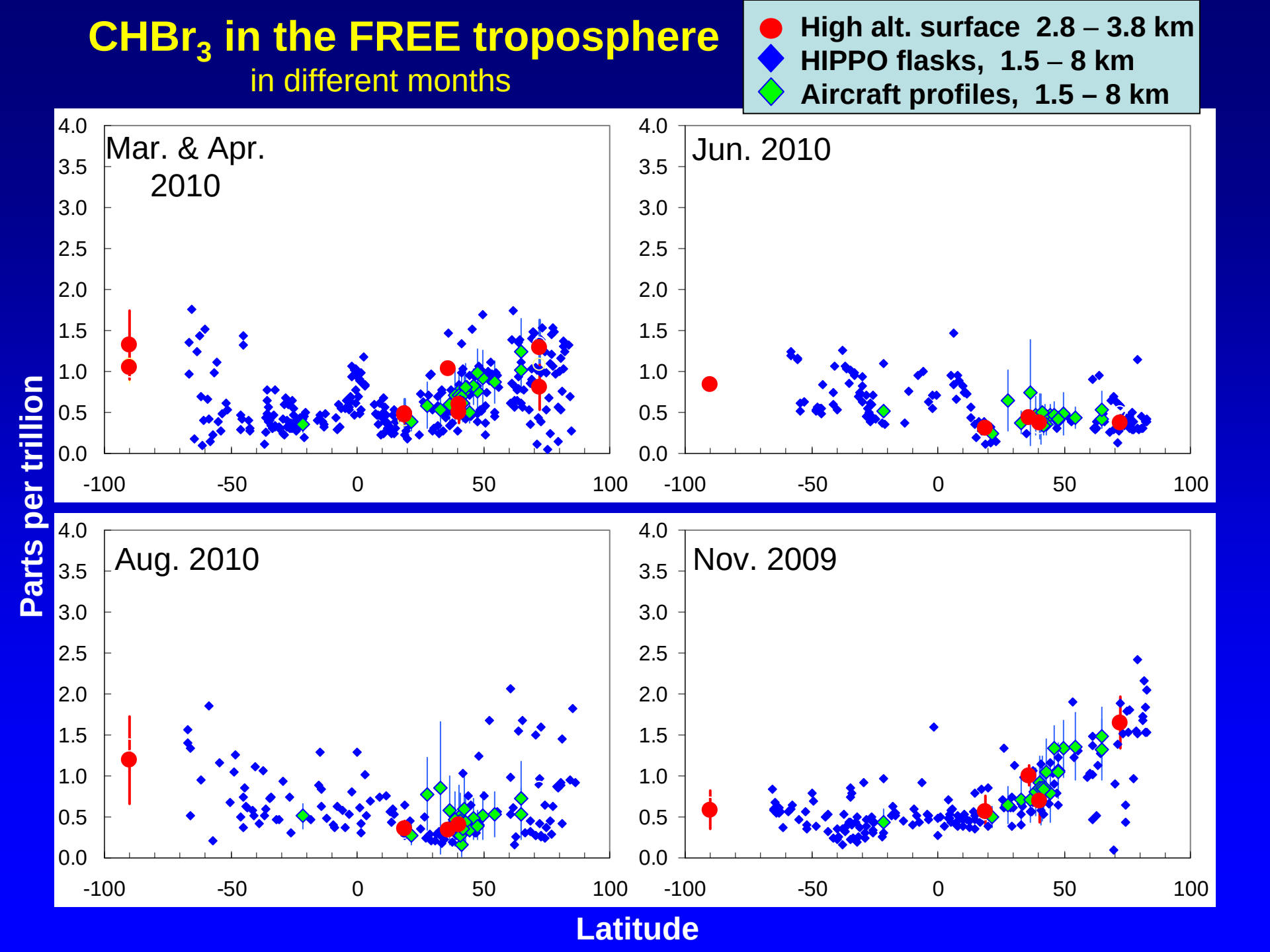
CH_2Br_2 in the troposphere

in different months



$CHBr_3$ in the FREE troposphere

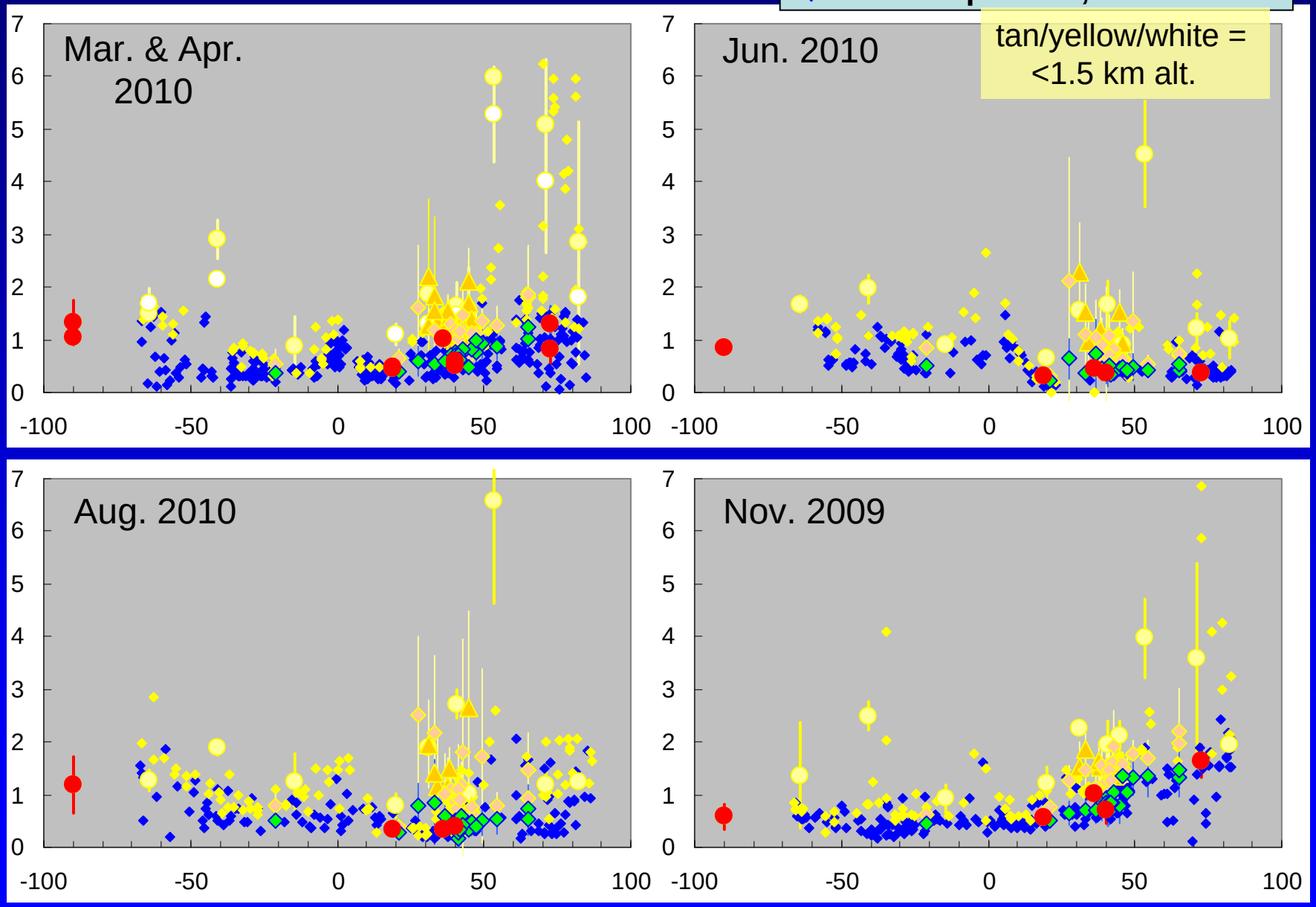
in different months



$CHBr_3$ in the troposphere

rescaled!

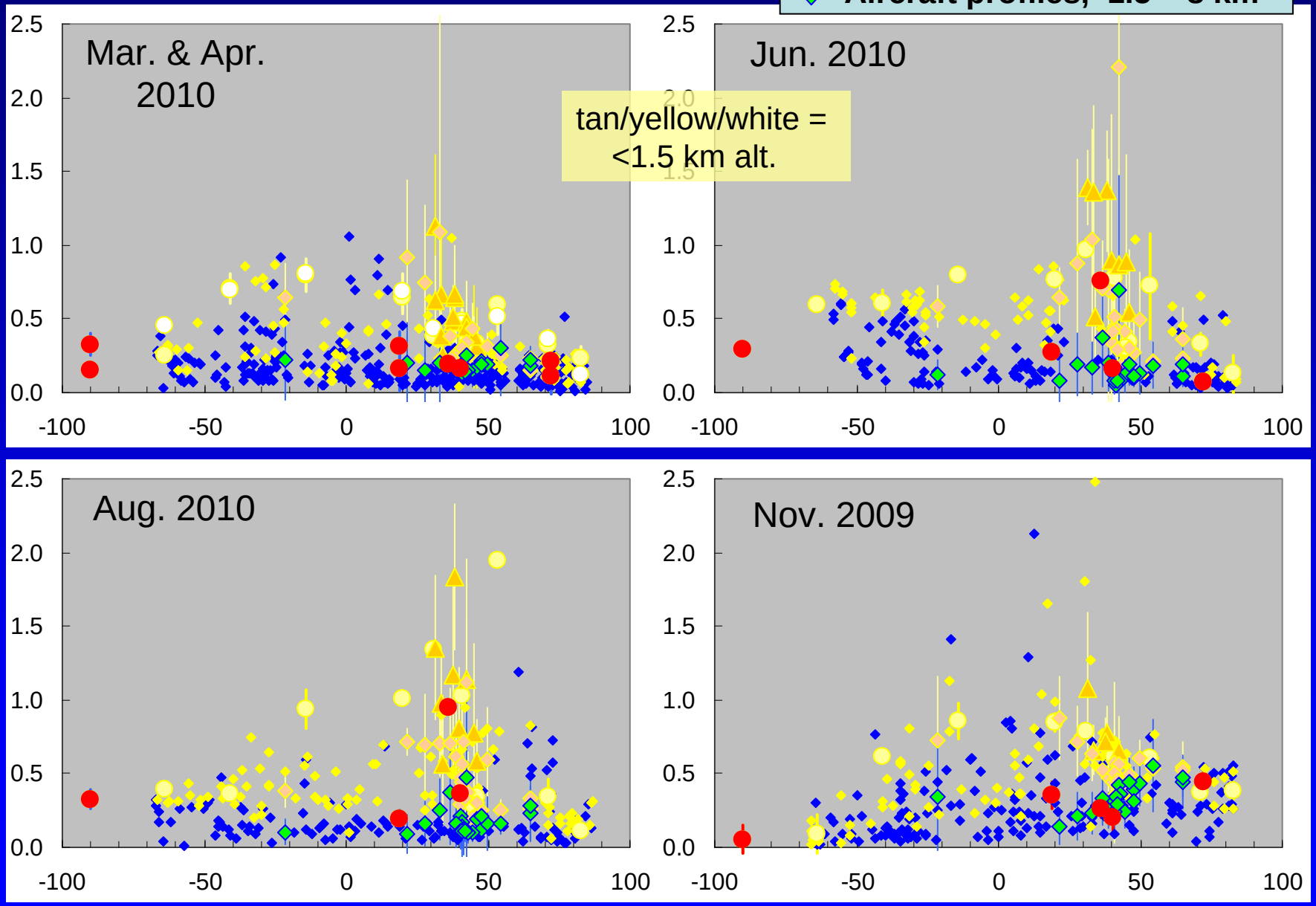
High alt. surface 2.8 – 3.8 km
HIPPO flasks, 1.5 – 8 km
Aircraft profiles, 1.5 – 8 km



CH_3I in the troposphere

in different months

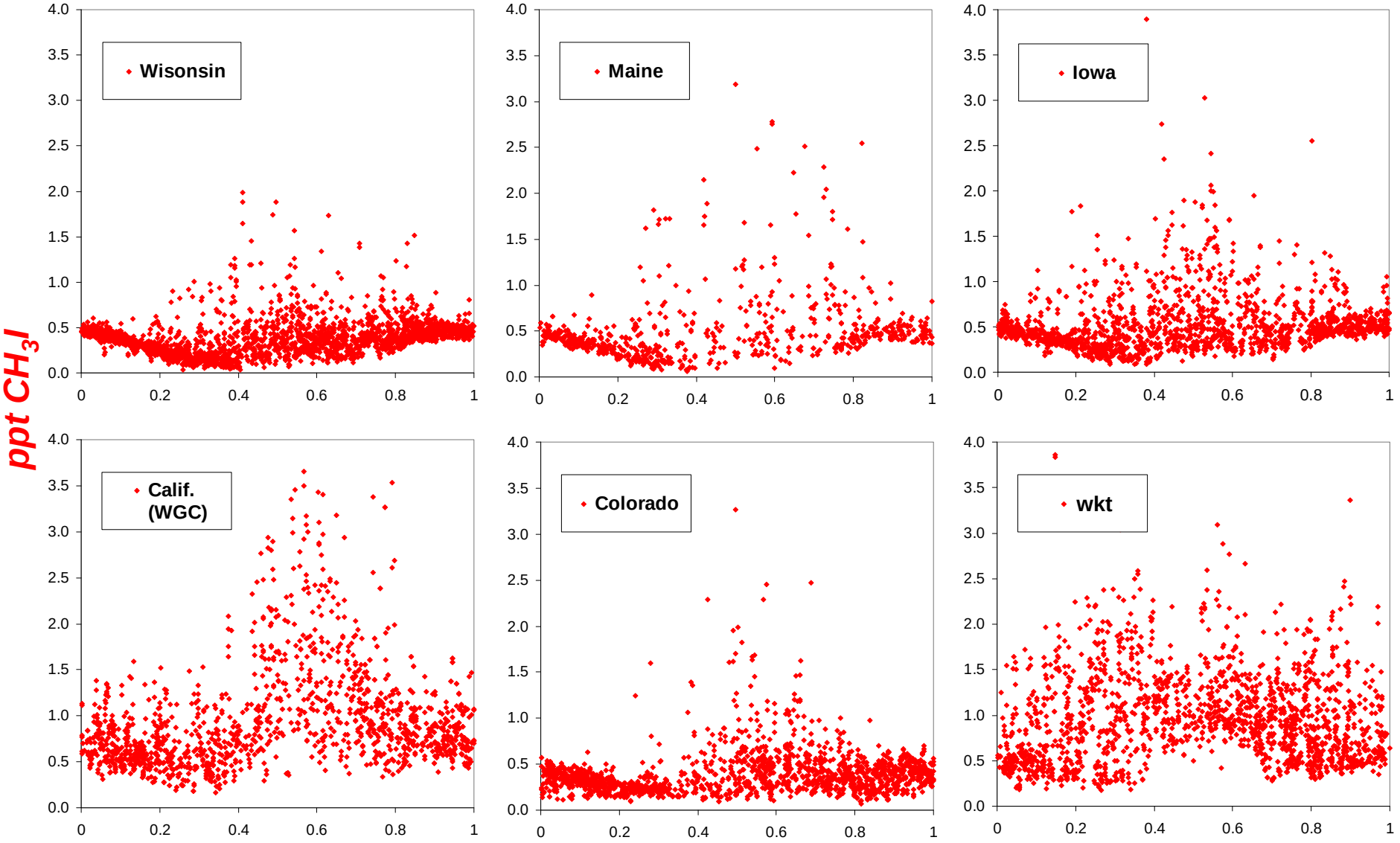
High alt surface 2.8 – 3.8 km
HIPPO flasks, 1.5 – 8 km
Aircraft profiles, 1.5 – 8 km



Parts per trillion

Latitude

CH_3I from selected TERRESTRIAL sites in the NOAA tower network



Summary:

Ongoing sampling and short-term projects allow us to characterize trace gas variability over latitudes, longitudes, and altitudes…

The results show:

→ Consistent mean mixing ratios from year-to-year for all three short-lived gases at a wide range of sites, but large site-to-site differences…

→ Higher consistency in results <u>from the free troposphere</u>… with seasonal variations driven primarily by losses especially for CH_2Br_2 and perhaps also $CHBr_3$. CH_3I exhibits higher variability (shorter lifetime!).

e.g., perhaps a true "background" for CH_2Br_2.

→ Boundary-layer mixing ratio enhancements above land *and* sea whose magnitudes vary seasonally (less so for CH_2Br_2, more so for $CHBr_3$ and especially CH_3I over land)

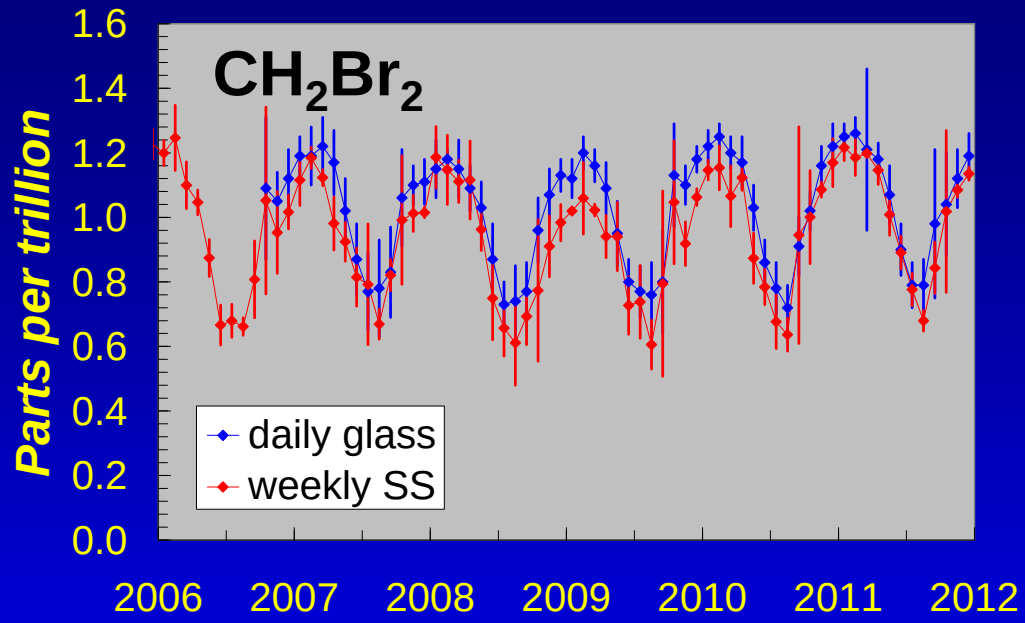
For short-lived gases, <u>free tropospheric data</u> provide an important context that allow an assessment of:

-distribution and seasonality of sources

-relative influence of local processes on any particular site

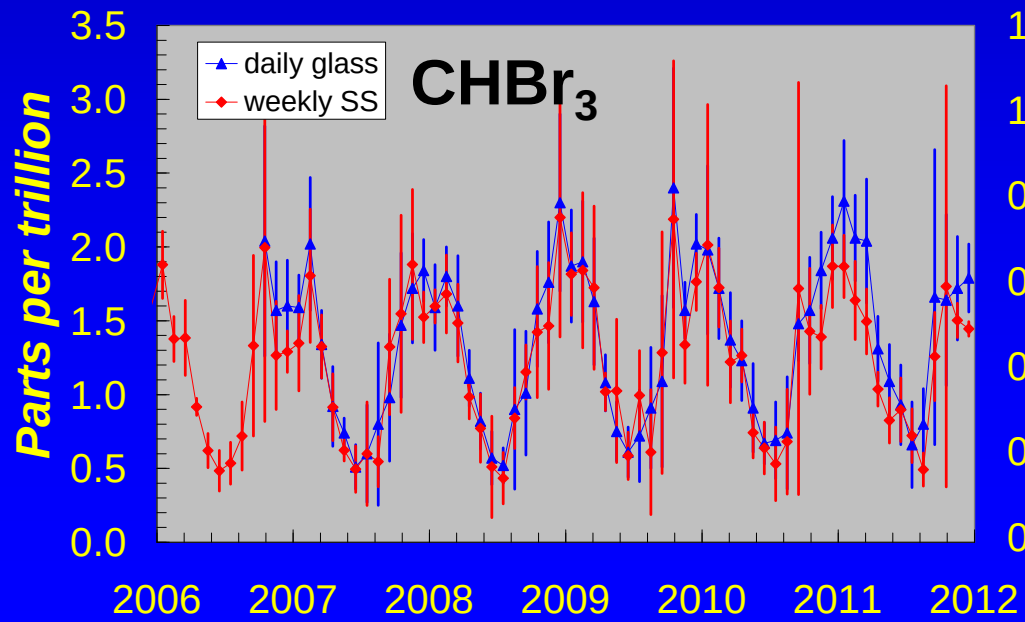
-usefulness of a site for monitoring long-term broad-scale changes

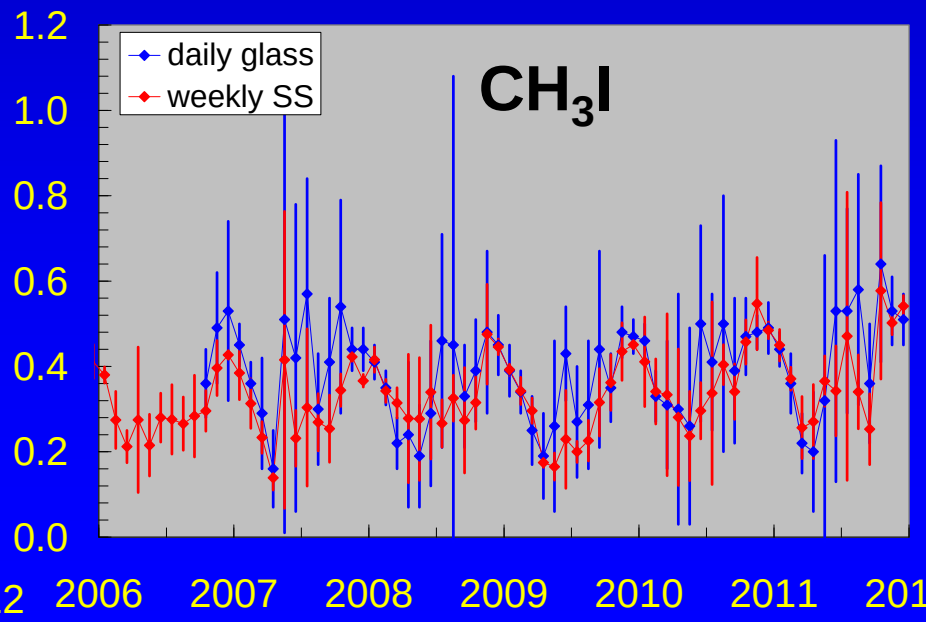
Glass (higher frequency vs SS (low freq) flasks at LEF (mid-west US) *…as monthly means*



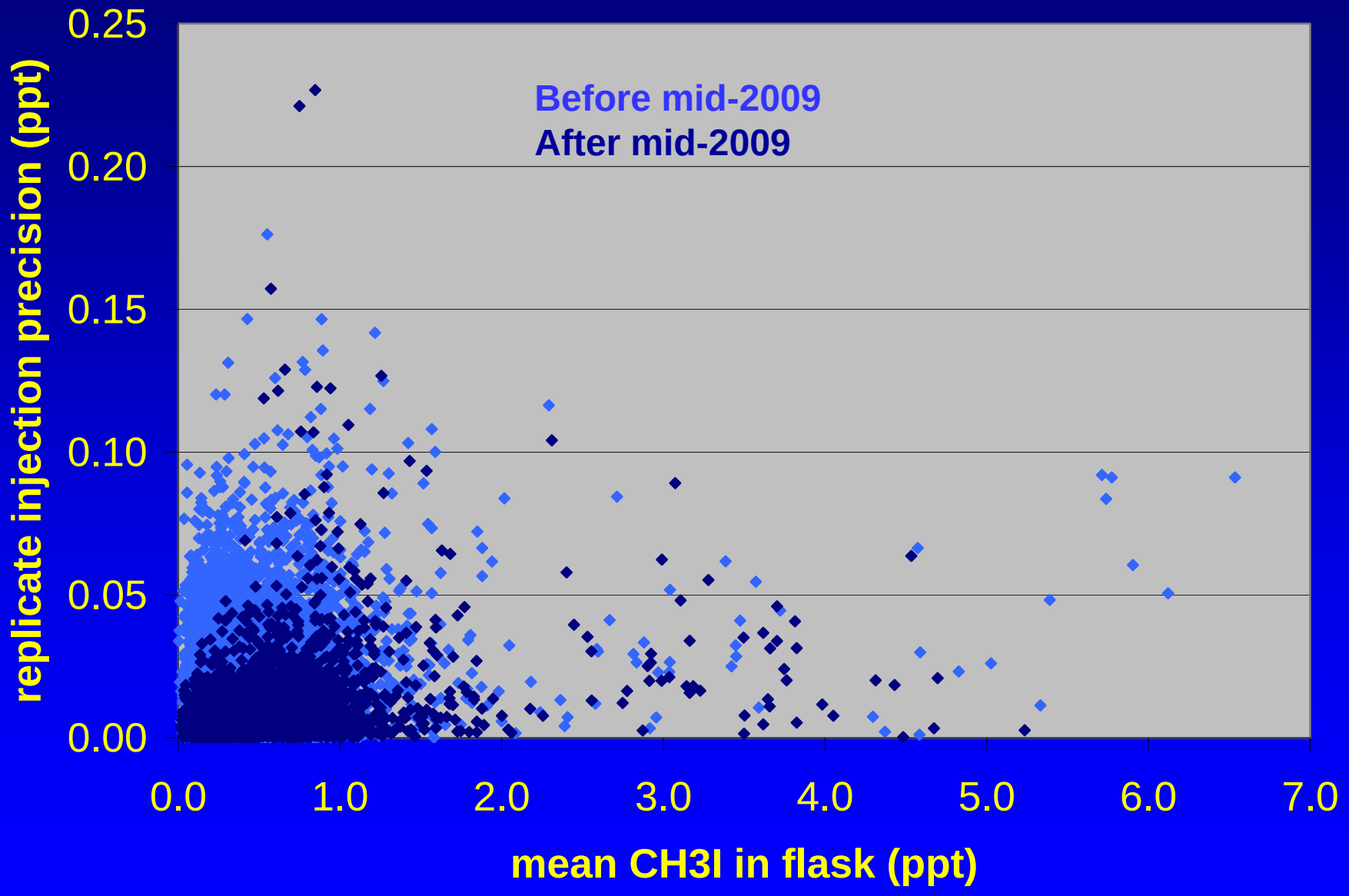
Plans to include:

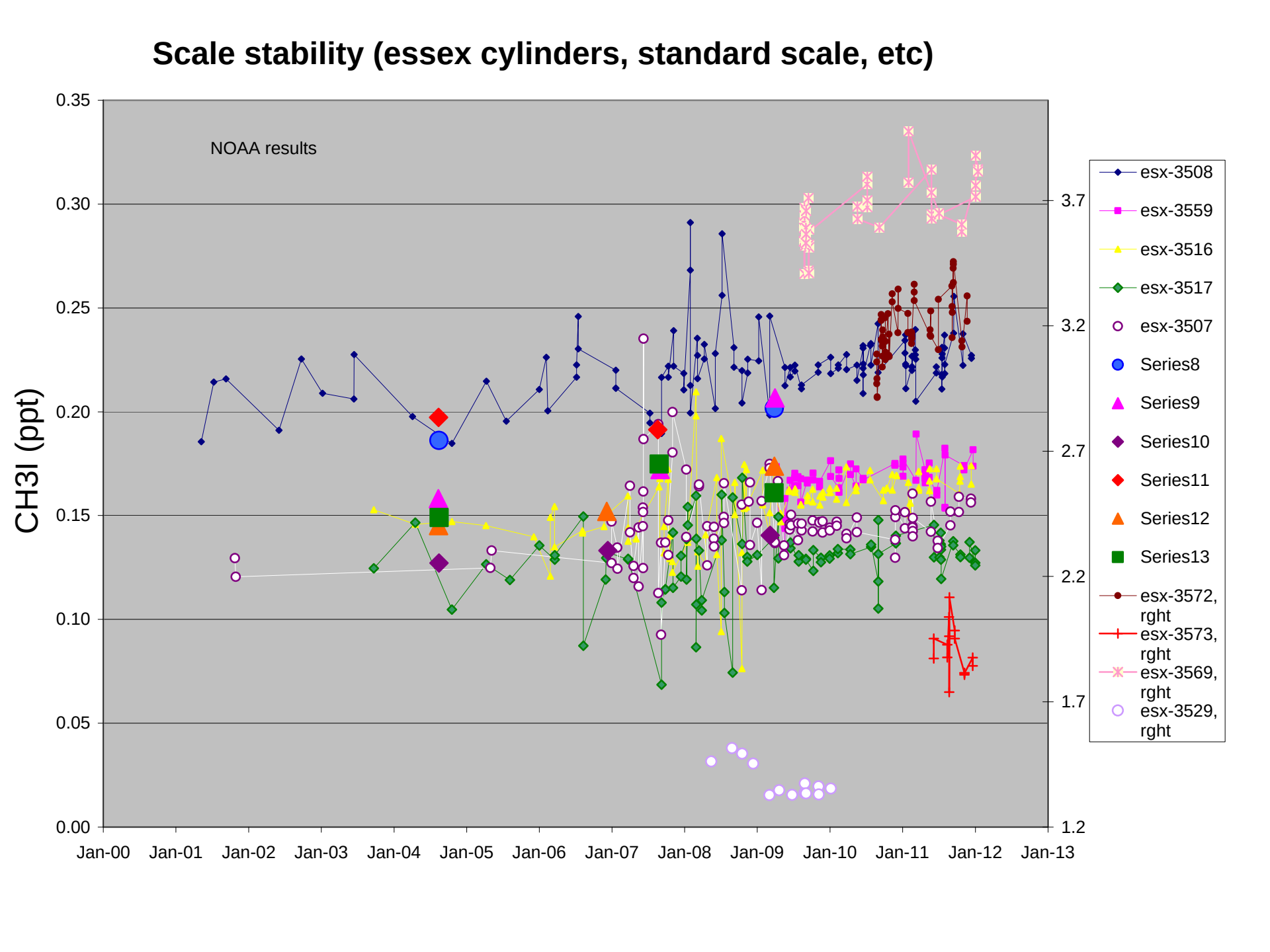
→ flask results vs. AGAGE *in-situ* instrumentation



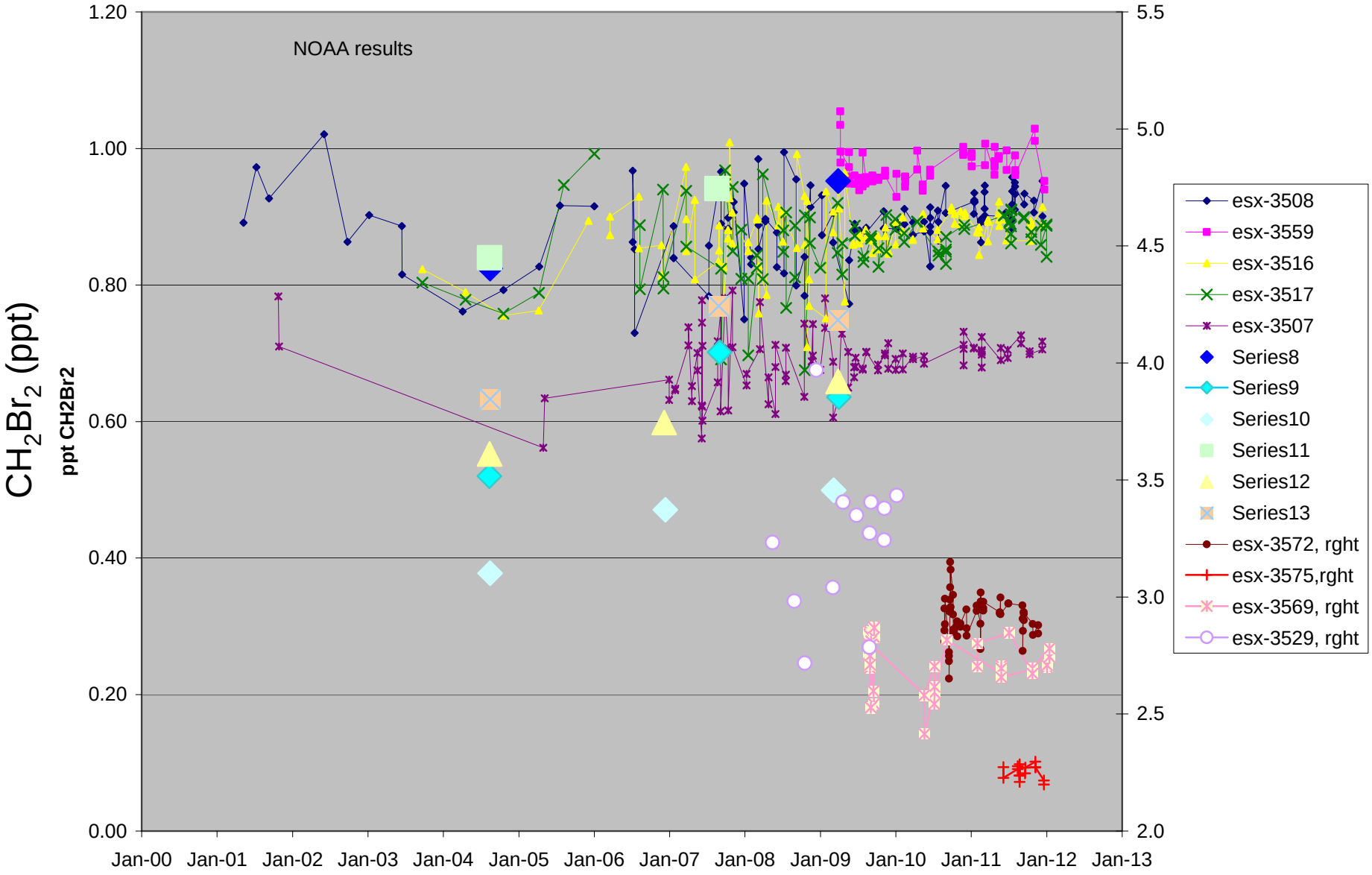


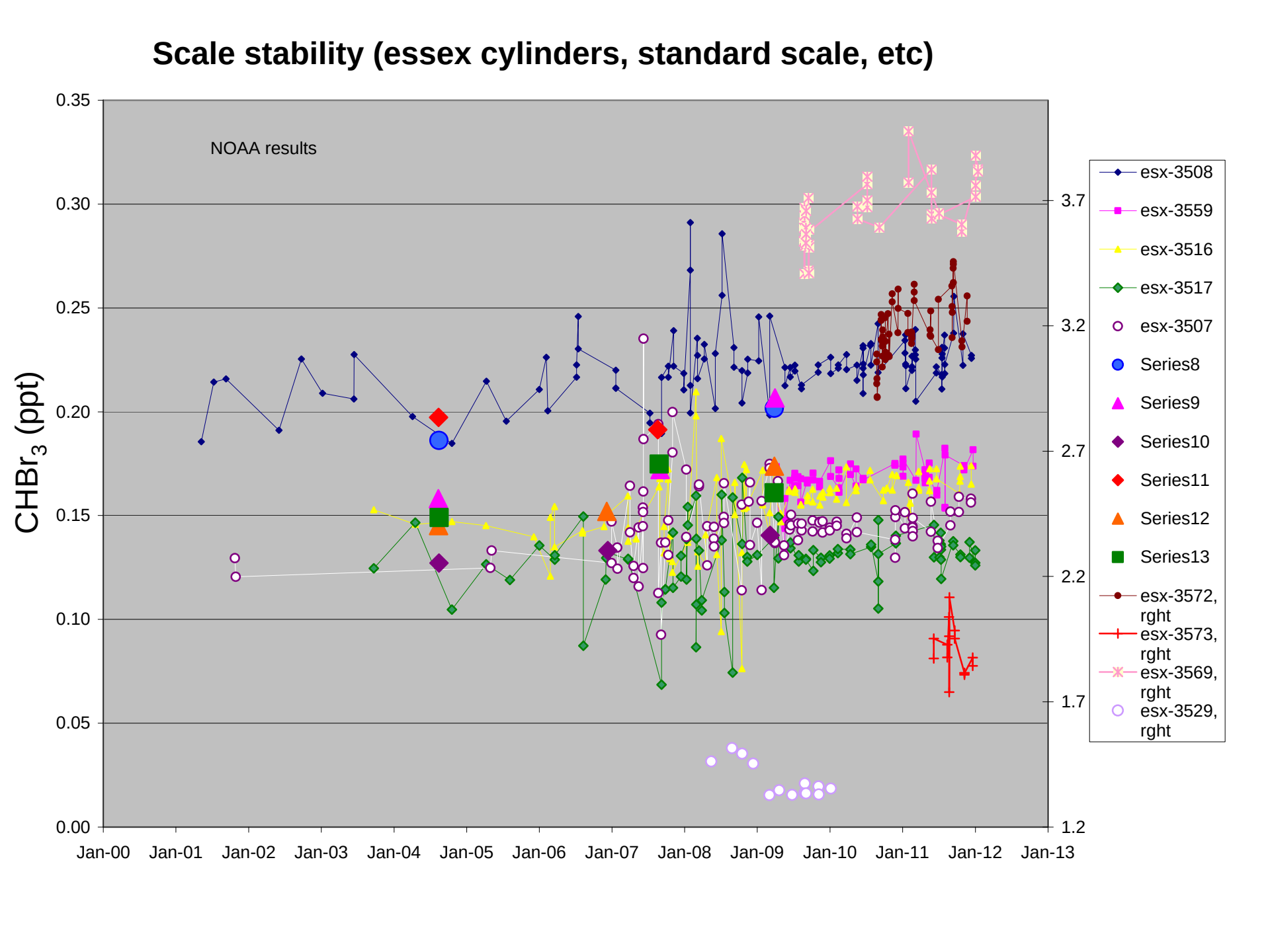
Measurement capabilities, methyl iodide




Scale stability (essex cylinders, standard scale, etc)
NOAA results
CH3I (ppt)
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
3.7
3.2
2.7
2.2
1.7
1.2
Jan-00
Jan-01
Jan-02
Jan-03
Jan-04
Jan-05
Jan-06
Jan-07
Jan-08
Jan-09
Jan-10
Jan-11
Jan-12
Jan-13
esx-3508
esx-3559
esx-3516
esx-3517
esx-3507
Series8
Series9
Series10
Series11
Series12
Series13
esx-3572, rght
esx-3573, rght
esx-3569, rght
esx-3529, rght

Scale stability (essex cylinders, standard scale, etc)




Scale stability (essex cylinders, standard scale, etc)
NOAA results
CHBr3 (ppt)
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
3.7
3.2
2.7
2.2
1.7
1.2
Jan-00
Jan-01
Jan-02
Jan-03
Jan-04
Jan-05
Jan-06
Jan-07
Jan-08
Jan-09
Jan-10
Jan-11
Jan-12
Jan-13
esx-3508
esx-3559
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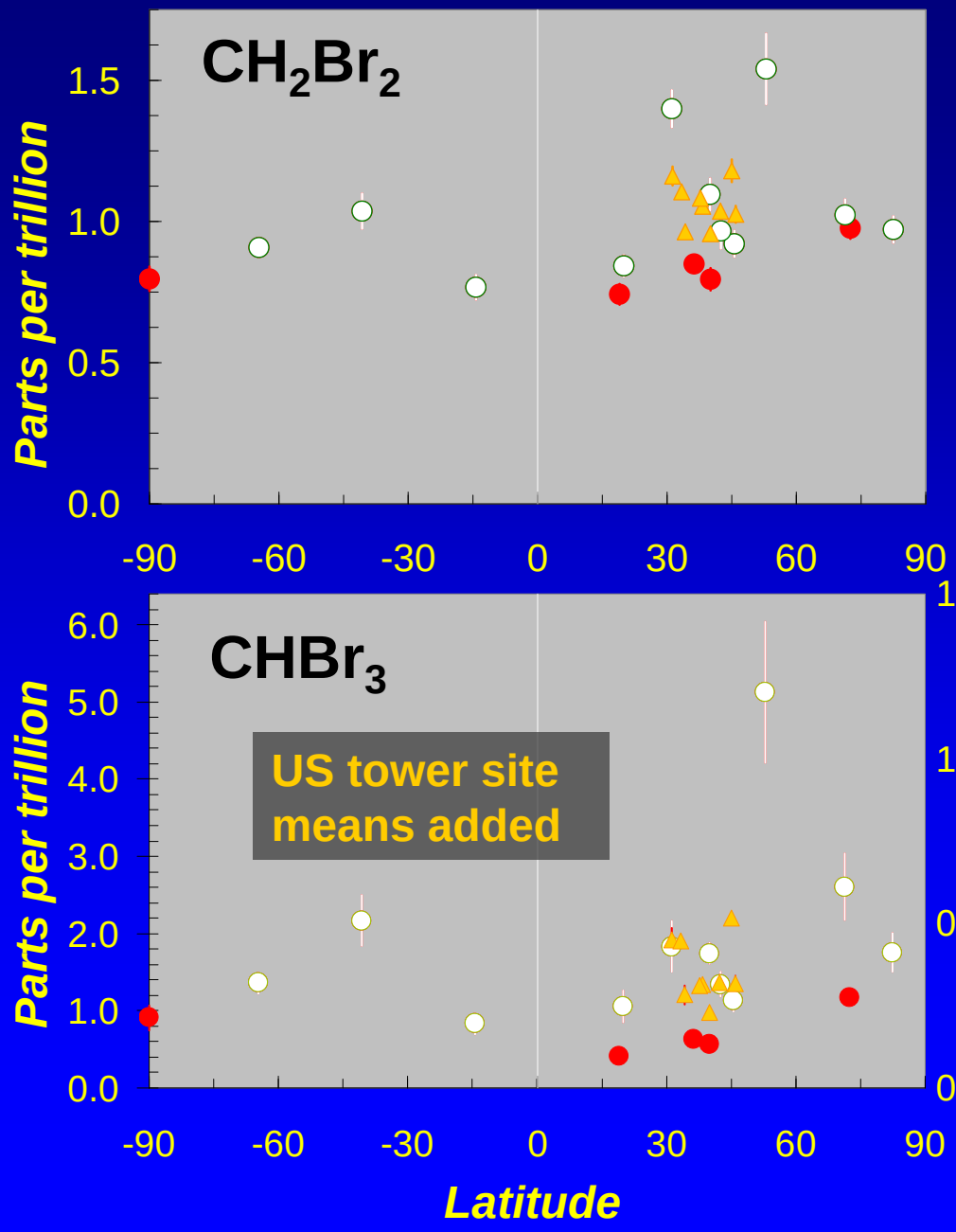
Annual means at surface sites *(2 to 17 yr records)*



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