Increases in tropospheric chlorine from dichloromethane, a gas not controlled by the Montreal Protocol.

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NOAA HATS flask results for dichloromethane (CH_2Cl_2) show large atmospheric increases in recent years:

Carpenter, L., S. Reimann, et al., WMO Ozone Assessment, 2014
Leedham-Elvidge, E.C., et al., Atmos. Chem. Phys., 2015
Hossaini, R., et al., Nature Geosci., 2015
Hossaini, R., et al., Geophys. Res. Lett., 2015, in press.

Why all the fuss?

CH_2Cl_2:

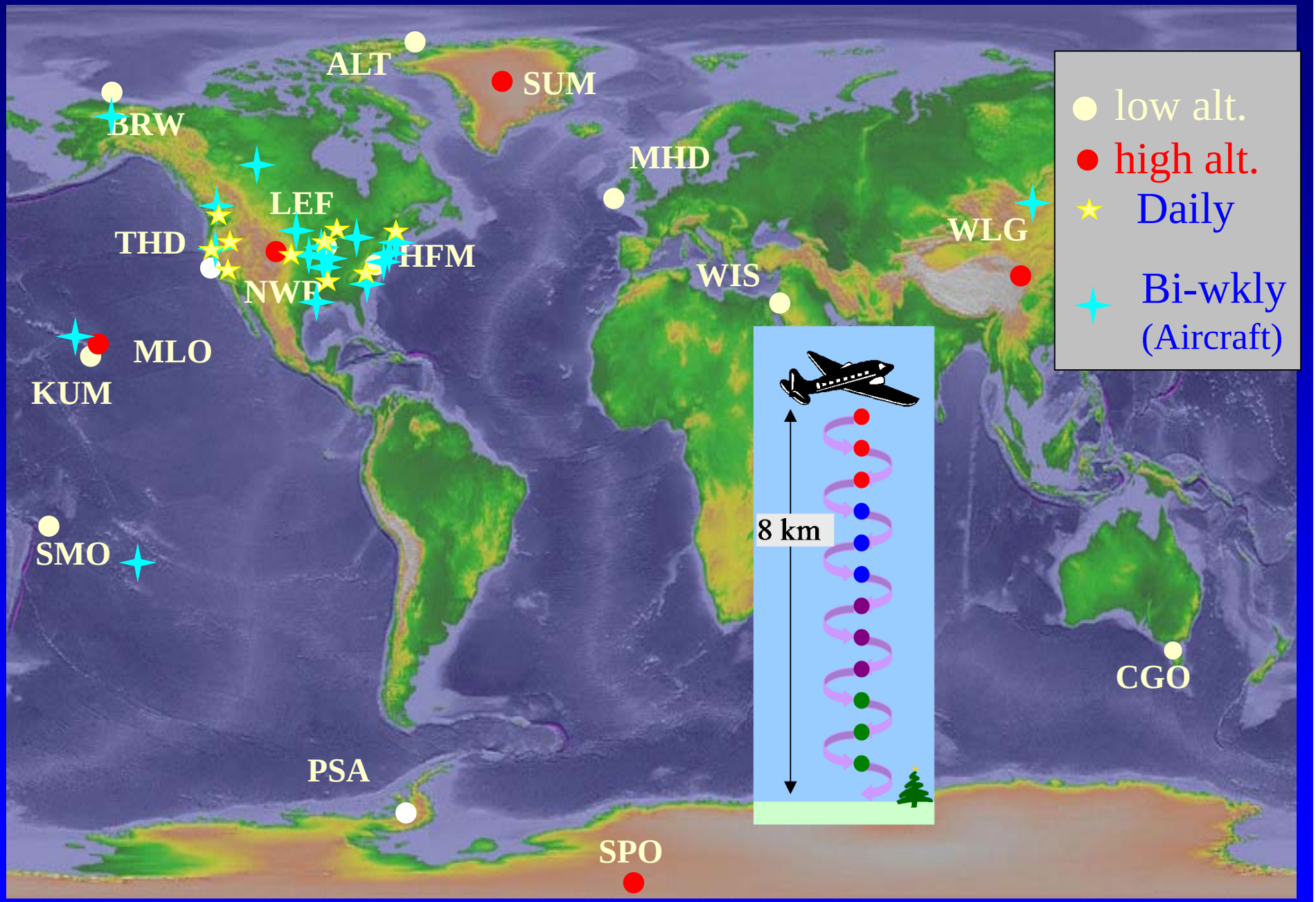
* *is emitted primarily from anthropogenic activities:*
  - *-solvent, cleaning agent, chemical reagent (HFC-32)*
  - *~800 Gg in 2012 (2 times Cl flux from F-12 or F-11 in the 1980s)*
* *is a short-lived gas (~5 month mean lifetime; 1.5 month in summer)*
* *ratio of [upper troposphere (TTL)] / [boundary layer] ~80%*

→ ***but is NOT controlled by the Montreal Protocol***

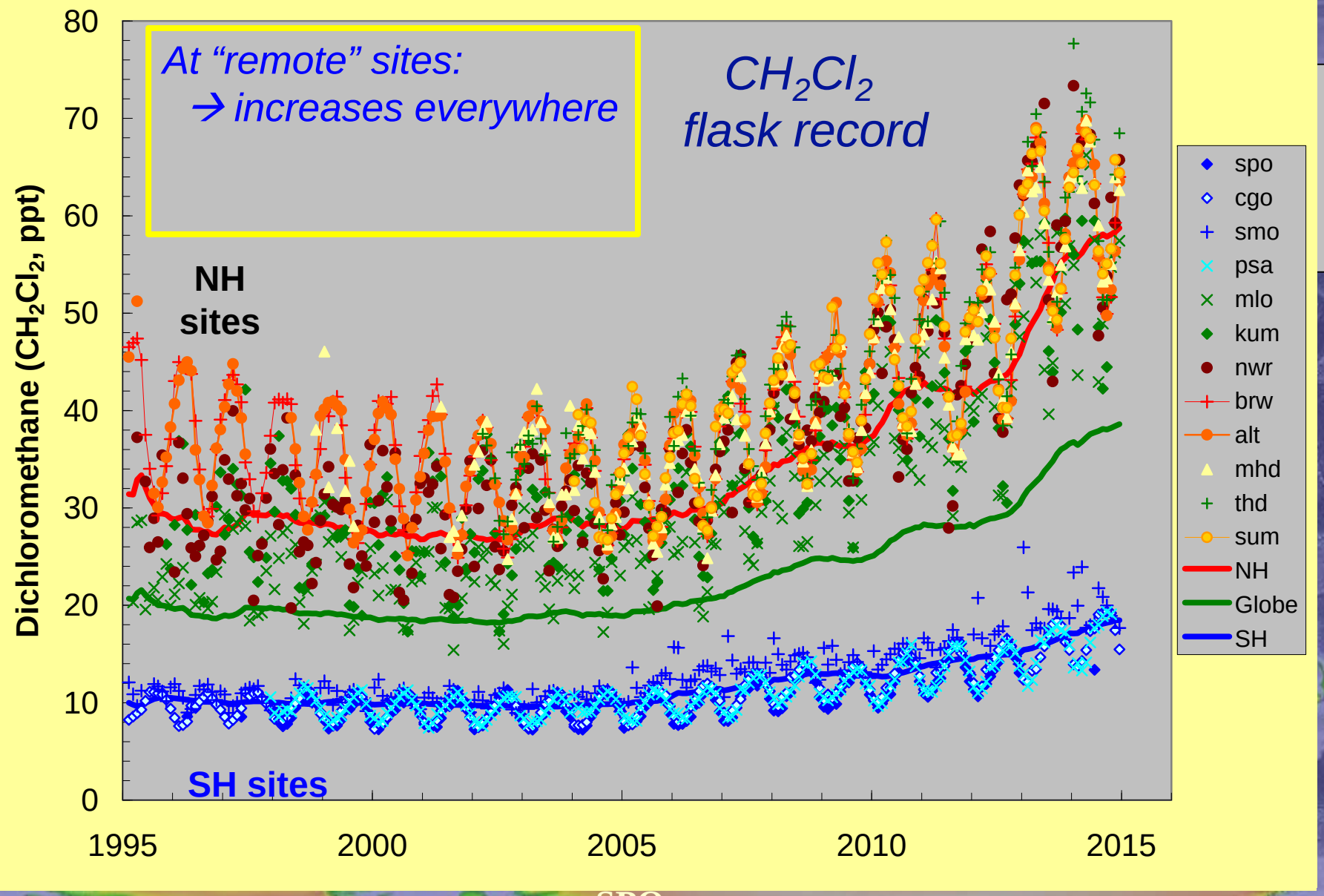
For today:

1) *how robust are changes observed for a short-lived gas?*
2) *how significant are changes for tropospheric chlorine?*
3) *where are the increased emissions coming from?*

The NOAA Halocarbon Sampling Network:



1a) How robust are the observed changes?



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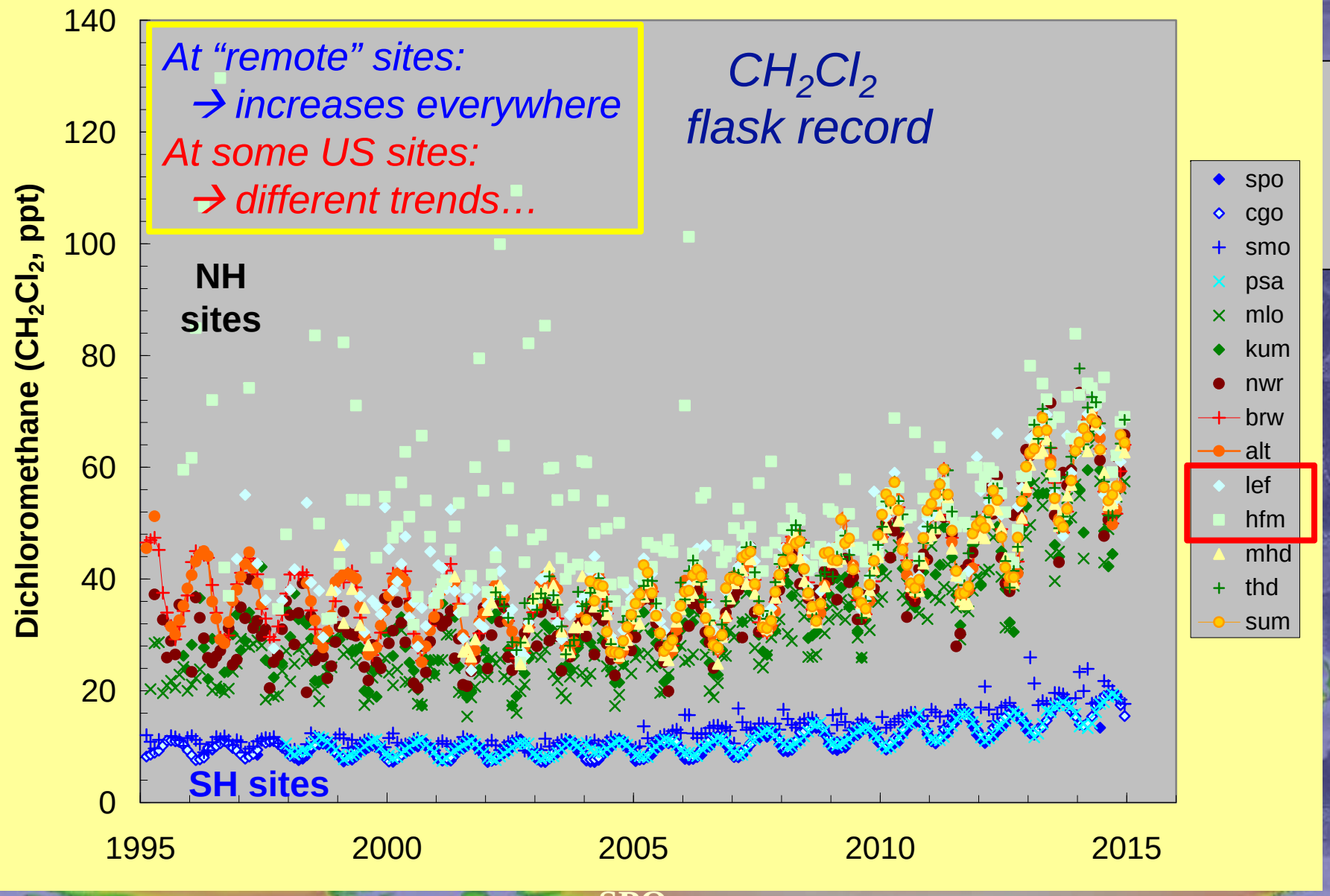


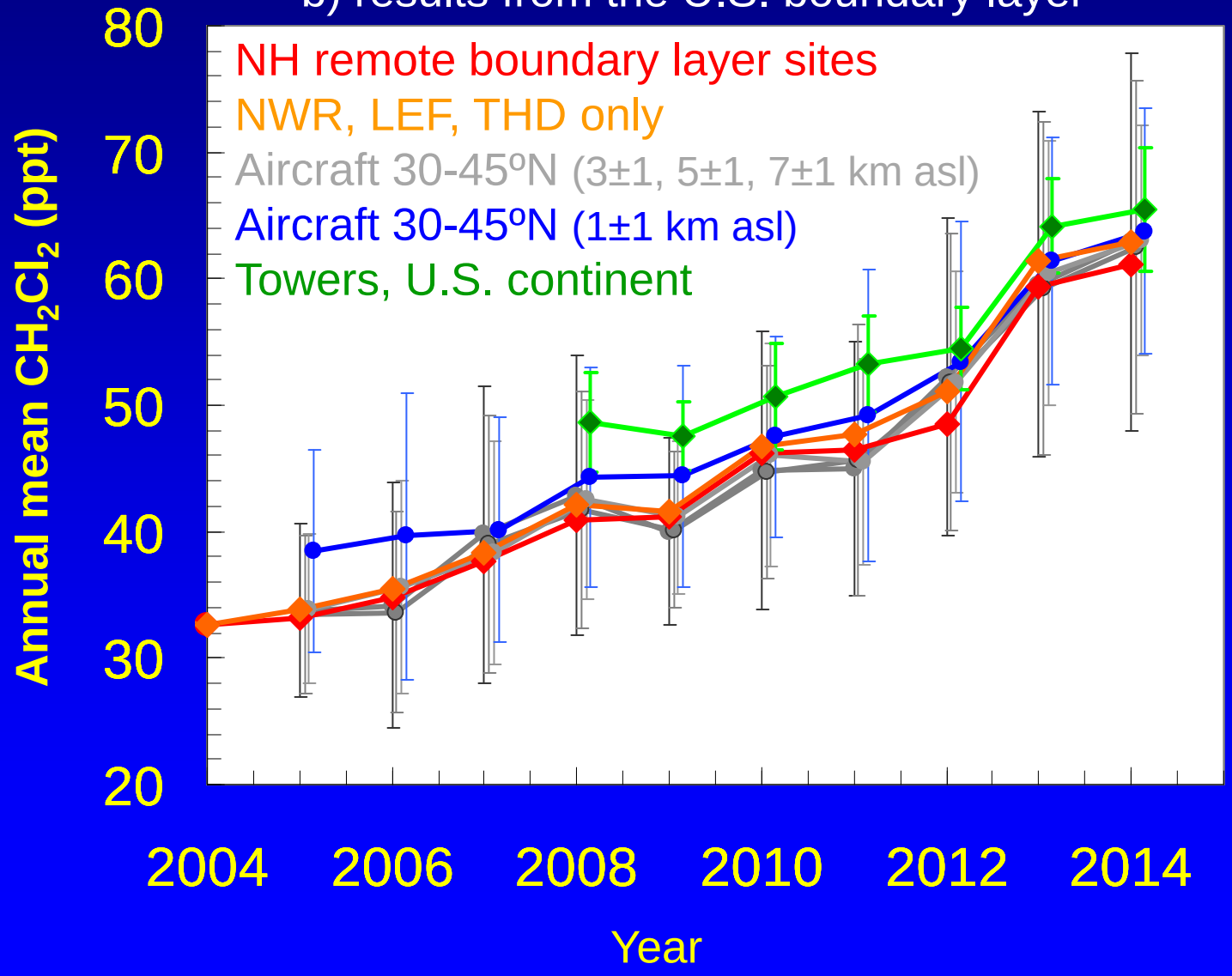
SPO

1b) How consistent are trends for a short-lived gas?

→ compare changes in 'remote' NH boundary layer to:

a) free troposphere means above the U.S.

b) results from the U.S. boundary layer



Changes at remote surface sites:

**** are consistent with those observed throughout the troposphere**

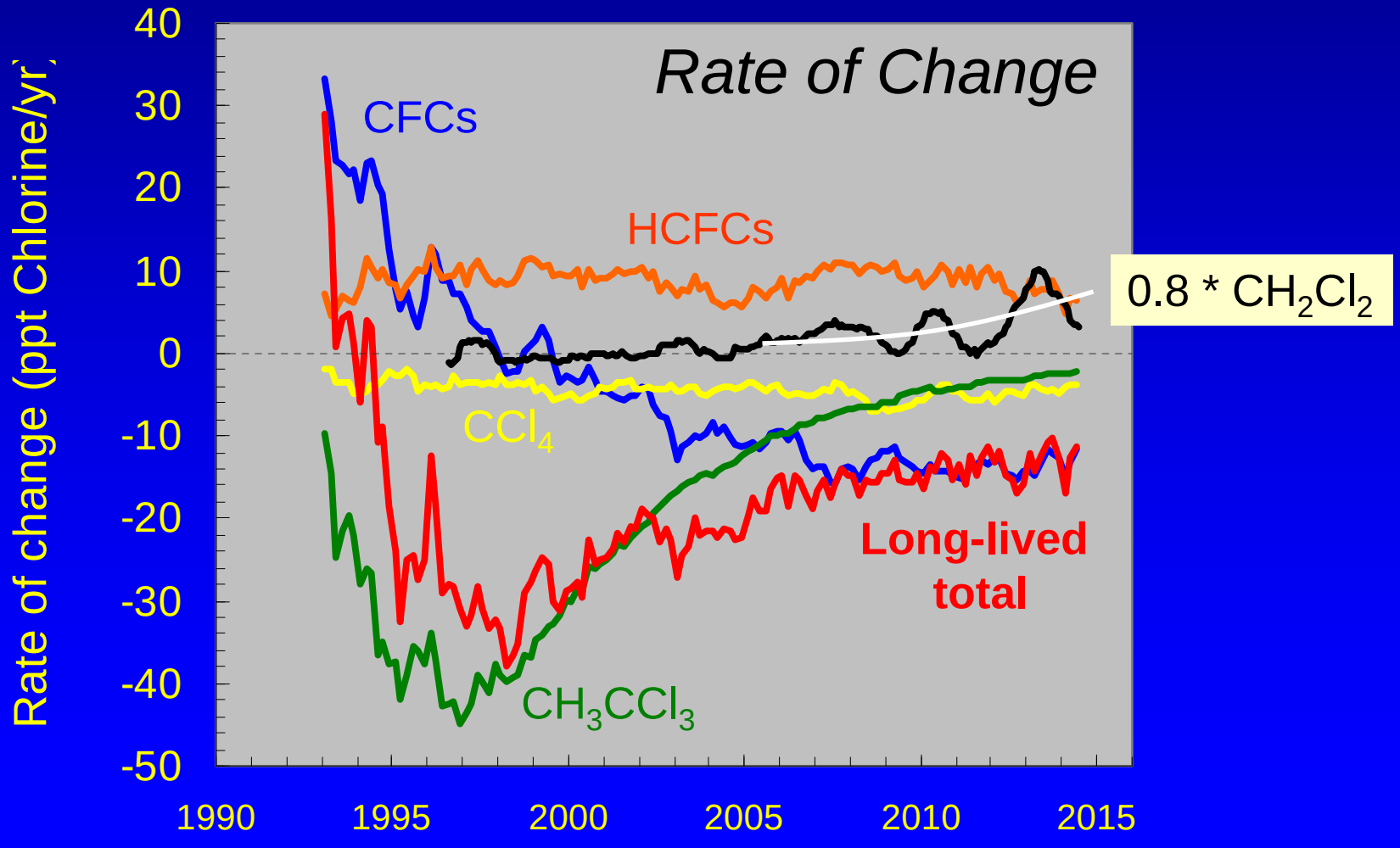
**** are larger than observed in the boundary layer over the U.S.**

NH remote bl sites:
KUM, NWR, THD, LEF, MHD, BRW, ALT

Towers sites:
AMT, BAO LEF, SCT, STR, WBI, WGC, WKT

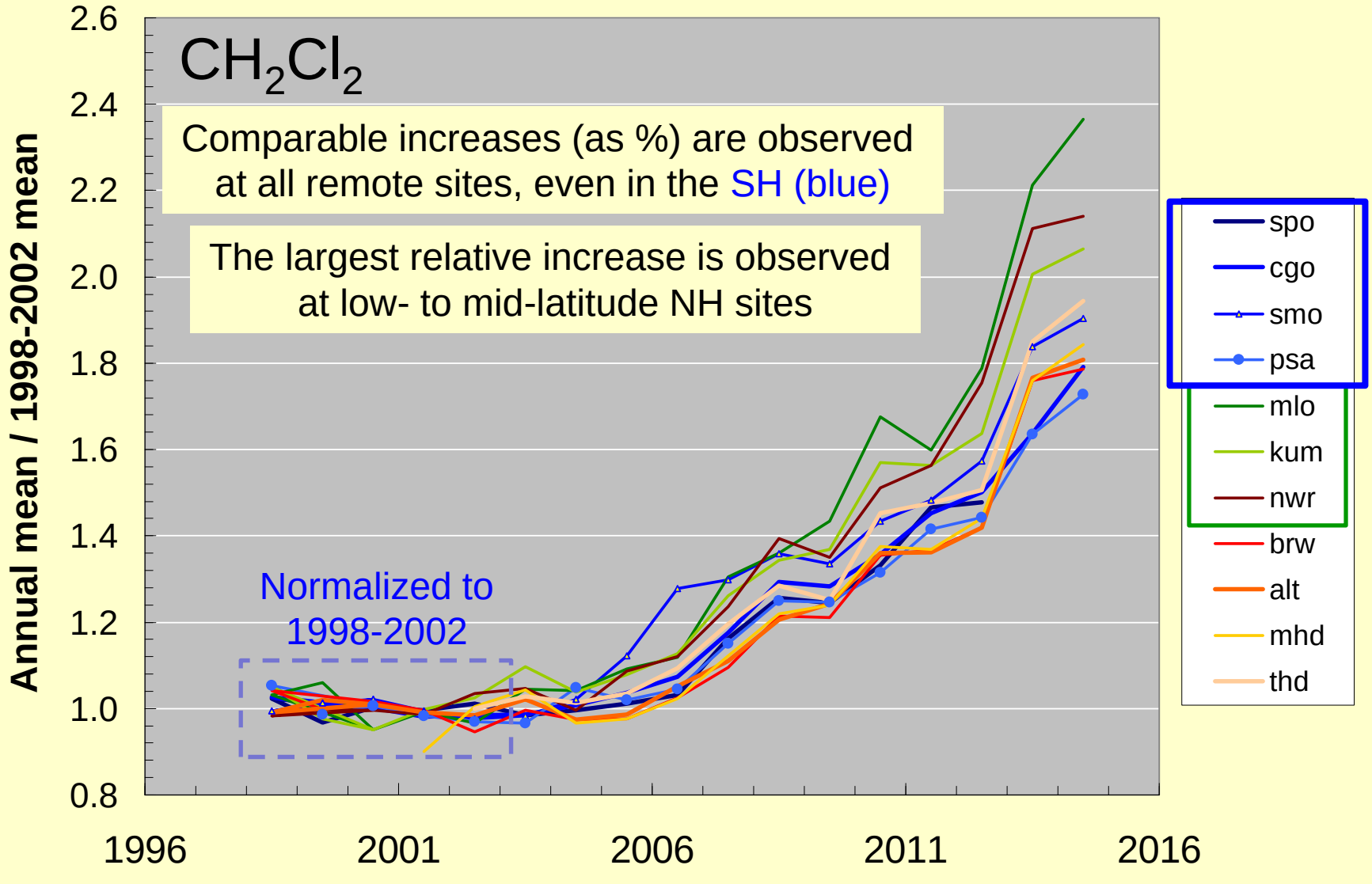
2) How large is the chlorine increase from CH_2Cl_2?

* 80 pptCl in surface CH_2Cl_2 means ~60 pptCl to the stratosphere
  → a larger contribution than HCFC-141b or HCFC-142b
* The rate of Cl increase from CH_2Cl_2:
  → is comparable to the Cl increase from the sum of all HCFCs



3a) Which latitudes are driving the increase?

The relative increase in annual mixing ratio by site:



3b) How have atmospheric distributions changed?

Intrahemispheric gradients:

NH: become smaller

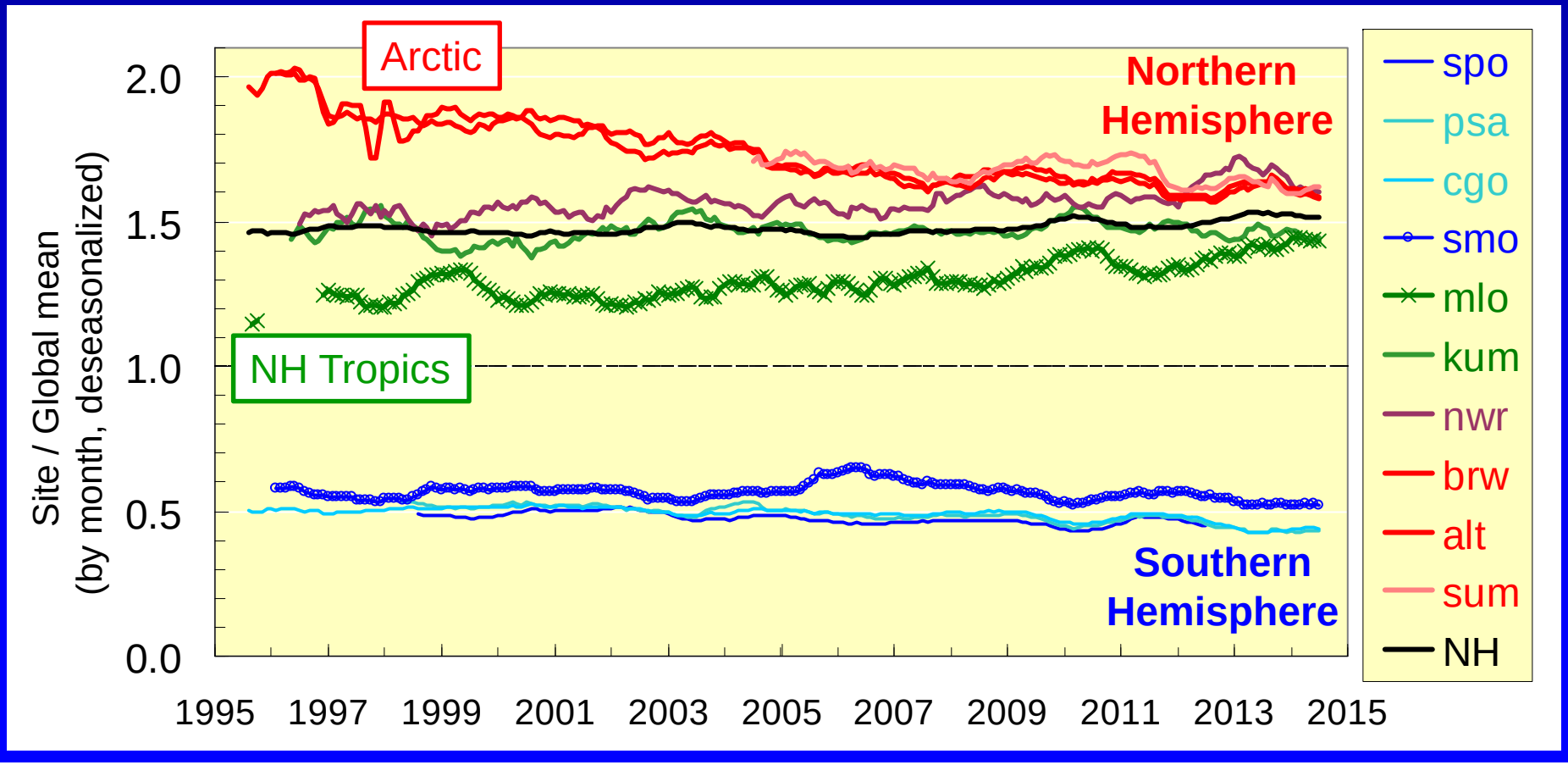
SH: slightly larger

→ NH emissions shifting to lower latitudes

Interhemispheric gradient:

Constant over time!

→ N vs S gradient set by time constants for loss and N – S exchange



Summary:

In flask results for CH_2Cl_2 since 1998-2002 we have observed:

*** consistent broad-scale changes in mole fractions (and seasonal variations) for a chemical with a 5-month global lifetime.**

Specifically:

*** about a factor of 2 increase at nearly all remote sites across the globe *and* consistent increases in the free troposphere above the U.S.**

*** reduced mole fraction enhancements in the U.S. boundary layer**

These imply:

*** substantial increases in global emissions, but not from the U.S. (U.S. emissions are likely decreasing)**

Changes in the observed atmospheric distribution imply:

*** a substantial shift in emissions to lower latitudes of the Northern Hemisphere**

Finally:

***stratospheric chlorine attributable to CH_2Cl_2 is currently *larger than contributed by either HCFC-141b or HCFC-142b* and is increasing at a rate *comparable to that from the sum of all HCFCs*.**